Nonlinear Viscoelastic Behaviour of HDPE

Constitutive Modelling and Finite Element Method Implementation
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PROEFSCHRIFT

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\( a_s(t_e) \) \hspace{1cm} \text{Time-ageing time superposition function}
\( a_r(T) \) \hspace{1cm} \text{Time-temperature superposition function}
\( a_s(\dot{e}) \) \hspace{1cm} \text{Time-strain superposition function}
\( a_o(\sigma) \) \hspace{1cm} \text{Time-stress superposition function (} a_{eq}, \ a_p, \ a_i \text{)}
\( B_0 \) \hspace{1cm} \text{Instantaneous bulk compliance}
\( C \) \hspace{1cm} \text{Compliance matrix}
\( d_y \) \hspace{1cm} \text{Deviatoric strain tensor}
\( D(t) \) \hspace{1cm} \text{Tensile creep compliance function}
\( D_o \) \hspace{1cm} \text{Instantaneous tensile creep compliance}
\( \Delta D(t) \) \hspace{1cm} \text{Transient tensile creep compliance function}
\( D \) \hspace{1cm} \text{Stiffness matrix}
\( E' \) \hspace{1cm} \text{Complex modulus}
\( E(t) \) \hspace{1cm} \text{Tensile relaxation modulus function}
\( E_o \) \hspace{1cm} \text{Instantaneous tensile relaxation modulus (modulus of elasticity)}
\( \Delta E(t) \) \hspace{1cm} \text{Transient tensile relaxation modulus function}
\( f \) \hspace{1cm} \text{Free volume fraction}
\( f_c \) \hspace{1cm} \text{Crystalline volume fraction}
\( g_o(\sigma) \) \hspace{1cm} \text{Schapery elasticity function (creep model)}
\( g_i(\sigma) \) \hspace{1cm} \text{Schapery pre-integral function (creep model)}
\( g_2(\sigma) \) \hspace{1cm} \text{Leaderman function, Schapery integral function (creep model)}
\( g_e(t_e) \) \hspace{1cm} \text{Strain-ageing time superposition function}
\( g_s(T) \) \hspace{1cm} \text{Strain-temperature superposition function}
\( \sigma \) \hspace{1cm} \text{Stress relaxation vector}
\( G_o(\sigma) \) \hspace{1cm} \text{Leaderman "excitation" function (creep model)}
\( G(t) \) \hspace{1cm} \text{Shear relaxation modulus function}
\( G_o \) \hspace{1cm} \text{Instantaneous shear relaxation modulus}
\( \Delta G(t) \) \hspace{1cm} \text{Transient shear relaxation modulus function}
\( h \) \hspace{1cm} \text{Thickness, height}
\( h_o(\varepsilon) \) \hspace{1cm} \text{Schapery elasticity function (relaxation model)}
\( h_i(\varepsilon) \) \hspace{1cm} \text{Schapery pre-integral function (relaxation model)}
\( h_2(\varepsilon) \) \hspace{1cm} \text{Leaderman function, Schapery integral function (relaxation model)}
List of symbols

- $H$: Enthalpy
- $H_2(e)$: Leaderman “excitation” function (relaxation model)
- $I_1, I_2, I_3$: First, second and third invariant of the stress tensor
- $I_2'$: Second invariant of the deviatoric stress tensor
- $J(t)$: Shear creep compliance function
- $J_0$: Instantaneous shear creep compliance
- $\Delta J(t)$: Transient shear creep compliance function
- $i, j$: Stress direction numbers
- $k$: Kelvin-Voigt element number
- $K_0$: Instantaneous bulk modulus
- $l$: Maxwell element number
- $M_e$: Number average molecular weight
- $M_w$: Weight average molecular weight
- $n$: Time point number
- $p$: Pressure
- $q(t)$: Value of the convolution integral (Schapery creep model)
- $r(t)$: Value of the convolution integral (Schapery relaxation model)
- $R$: Boltzmann constant 8.314 J.mole$^{-1}$.K$^{-1}$
- $s_{ij}$: Deviatoric stress tensor
- $t$: Time
- $t_c$: Time from the start of creep
- $t_r$: Time from the start of recovery after creep
- $t_s$: Time from the start of stress relaxation
- $t_e$: Ageing time
- $T$: Temperature
- $T_g$: Glass transition temperature
- $T_m$: Melt temperature
- $U$: Energy
- $V$: Volume
- $\alpha$: Stepping parameter
- $\alpha$: Thermal expansion coefficient
- $\gamma$: Shear strain
- $\gamma_{eq}$: Equivalent strain
- $\delta$: Phase lag
- $\delta_{ij}$: Kronecker delta
- $\varepsilon$: Strain
- $\varepsilon_c$: Strain during creep
- $\varepsilon_r$: Strain during recovery after creep
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<tr>
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<tr>
<td>$\varepsilon_{vp}$</td>
<td>Viscoplastic strain</td>
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<tr>
<td>$\varepsilon_m$</td>
<td>Volumetric strain</td>
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<td>$\varepsilon_{ij}$</td>
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<td>Stress tensor</td>
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<td>Stress tensor</td>
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<td>$\tau$</td>
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1 Introduction

1.1 Background

It is mostly the relatively low cost and large freedom in production of complex shapes by injection moulding which have resulted in a large variety of structural applications of plastics. Furthermore the low density of polymers enhances the design of lightweight products. The relatively low stiffness of polymers however reduces its applicability for products.

Weight reduction of products is becoming increasingly important because of cost reduction and environmental benefits. As a consequence the material will have to perform in its outer limits of its mechanical capability. Consumer products are often designed with plastics and consequently thorough knowledge of the mechanical behaviour of this class of materials is essential.

The time dependent mechanical behaviour of polymers complicates the design of products with minimal weight. The mechanical response of complex shaped plastic products can be evaluated numerically with Finite Element Method (FEM) calculations, if only the material behaviour is modelled correctly. However the nonlinear time dependent behaviour of polymers is mostly approximated resulting in loss of accuracy. FEM calculations with nonlinear time dependent material behaviour are scarce and therefore plastic products are not seldom overdesigned using more material than strictly needed.

The time dependent mechanical behaviour of plastics is also called viscoelasticity. If a viscoelastic material is loaded with a constant load then the deformation will increase in time, corresponding to a lower stiffness. This is called creep. The process in which after unloading the original material configuration is regained in time, is called recovery. In case of a constant deformation the decay of stiffness in time will reveal itself in a decay of the stress. This is called stress relaxation.

A typical example of viscoelastic behaviour of a plastics consumer product can be found in the figure 1.1. A stack of bottle crates is shown. Because of the weight, especially the lower crate will creep and the decreasing stiffness might even cause buckling. Another practical problem, for example, is creep in pressurized pipelines for drinking water.
Bottle crates such as displayed above, containers and many other products are composed of High Density PolyEthylene (HDPE). This polymer exhibits a marked viscoelastic behaviour. Because of creep the deformation of this material can easily double in time, even at relatively small load levels, which may occur during service of a product. This material is chosen for further investigation, since weight reduction for these products with HDPE is plausible when exact mechanical responses can be calculated.

Polymers are formed during a polymerization reaction when monomers are coupled to long chainlike molecules. The chemical structure of the chains defines the material. However within that material class a large variety of properties can be accomplished by variation of the chain length distribution and the amount of branching of this chain, i.e., physical structure.

HDPE is a thermoplastic, meaning that the long chain-like molecules are not interconnected by chemical cross-links and so the material will melt when heated. However the viscosity of the melt is very high due to the entanglements between the molecules, which act as physical cross-links. The larger the molecules, the more difficult these entanglements can be disrupted and the higher the melt viscosity.

Two types of thermoplastics can be distinguished: amorphous and semi-crystalline thermoplastics. Amorphous thermoplastics solidify when cooled below the glass transition temperature due to the decreasing mobility of the polymer chains. Semi-crystalline
thermoplastics, such as HDPE, become solid at the melt temperature when the chains are forming crystals. Part of the polymer chains however do not crystallize and will remain amorphous resulting in a composite structure. When the temperature is below the melt temperature and above the glass transition temperature the amorphous segments will be very flexible. This is the case for HDPE, which has a melt temperature around 90°C and a glass transition temperature of approximately -115°C. Cooled below the glass transition temperature even these mobile segments solidify and the material will become stiffer.

The degree of crystallinity and the structure of the crystals not only depend on the physical structure of the molecules, such as degree of branching, but also depend on the thermal history. The cooling rate after injection moulding of HDPE products therefore determines the mechanical behaviour, since a higher degree of crystallinity is accompanied by a higher stiffness.

Since viscoelasticity mostly manifests as creep during service of a product, the characterization of HDPE was chosen to be done in terms of creep as well. Furthermore creep experiments are easily performed and reproduced, and constant loads with long time spans can be accomplished by means of a dead weight. Also during creep the complete stress state is known whereas during a stress relaxation experiment the complete strain state is not known, a combined stress/strain situation appears.

![Figure 1.2](image)

Figure 1.2  Approach to describe viscoelasticity.

Choosing the tensile creep experiment on HDPE as a reference measurement, different quantities can be altered. To retain order in the treatment of viscoelasticity and to reduce the number of experiments to acceptable levels, it is chosen that only one single quantity is to be changed at a time. This approach is visualized in figure 1.2. For instance the effect of
temperature on creep is investigated as well as the effect of the time after production of the specimens, i.e., the age of the material. But both effects are not addressed simultaneously.

The reference for all measurements is uniaxial tensile creep of HDPE 7058Z at a temperature of 23°C and a material age of 32 weeks. For further information see chapter 3. The effects to be studied are:

**Arbitrary stress histories.** Different stress histories such as recovery after creep, tensile straining or stress relaxation are investigated.

**Temperature.** The influence of temperature on creep is investigated.

**Ageing.** The effect of the material age on the creep behaviour is considered.

**HDPE grade.** Three types of HDPE were tested for their creep response.

**Non-tensile stress states.** Tests with non-tensile stress states like compression, torsion and bending are to be realized.

The effect of the production process on the material properties is excluded in this research.

After the characterization of mechanical behaviour of HDPE with existing models, the most suitable material model is applied for FEM analyses. A robust and fast numerical treatment of the material model is developed for FEM calculations. Therefore different time integration strategies are programmed. Some of these are equivalent to those already in use for analyses with plasticity. The feasibility of complex calculations with FEM is evaluated by a case study on the response of an HDPE bottle crate. Both the numerical stability and the computation times are investigated for the different numerical methods.

The creep response of plastic products is often evaluated with simplified FEM calculations. The material behaviour at a certain creep time is then approximated by the (nonlinear) stress/strain relation found by the creep curves at that creep time, i.e., creep isochrones. The validity of the simplified calculations is verified for creep, stress relaxation and straining with constant strain rate.

### 1.2 Objectives

An accurate description of the mechanical behaviour of plastics and the ability to calculate the response of complex structures are required for the design of plastic products with minimal weight. Therefore the two main objectives of this research are the description of the viscoelastic behaviour of polymers and the implementation of the material model into a
1.2 Objectives

FEM package for numerical analyses of products.

The semi-crystalline polymer HDPE was chosen for this research, because of its wide use in products like bottle crates and containers and its marked viscoelastic behaviour. Its creep behaviour is modelled with various existing nonlinear viscoelastic models, which are known from literature. The models are compared in terms of ability to describe the HDPE data, ease of handling, and also its generality, i.e., its ability to describe polymers other than HDPE.

In order to develop a robust numerical scheme for FEM calculations, various time integration strategies of the most suitable nonlinear viscoelastic model are programmed into a FEM package and compared for their numerical stability and computation time. Furthermore the validity of simplified analyses using creep isochrones is verified.

1.3 Outline

This thesis starts with a literature survey on time dependent behaviour of polymers in chapter 2. Different models which are used to describe viscoelastic behaviour are explained, and the numerical tools needed for calculations are derived.

Chapter 3 discusses the experimental procedures. First the creep experiments on HDPE are described followed by the experiments with an arbitrary stress history such as creep recovery, tensile straining, ramp loading and stress relaxation. Also creep experiments with different temperatures, ageing times and grades of HDPE are described. Chapter 3 ends with a description of the creep experiments with multiaxial stress states.

The modelling of the experimental results will be addressed in chapter 4. First the creep results before yielding are described using the Schapery creep model. Next the yield behaviour is included with a model with two parallel processes. Also the Schapery model in relaxation form can describe the behaviour up to yielding. All three models are used to predict the strain during arbitrary stress histories. A viscoplastic term is added to the Schapery creep model to describe the recovery behaviour. Finally the effects of temperature, ageing, and the HDPE grade are examined.

The implementation of the Schapery creep model into the FEM package MARC is described in chapter 5. First the uniaxial model is extended to multiaxial stress situations. Then different numerical schemes are described and compared. The experimental results of creep with non-tensile stress states are compared with the FEM predictions. Finally in a study on
the viscoelastic response of an HDPE bottle crate full viscoelastic analyses are compared with simplified analyses with creep isochrones. The thesis is concluded with the consequences of this research for designers, conclusions and recommendations.
2 Time dependent mechanical behaviour

2.1 Introduction

Thermoplastics exhibit a pronounced time dependent mechanical behaviour. This behaviour implies that a unique stress strain relation is not present, but that the current strain is determined by the complete stress history. Likewise the current stress is determined by the complete strain history. The material is said to have a memory.

Time dependent behaviour manifests itself when a constant load induces a deformation which is increasing in time. This is called creep. The term stress relaxation represents a decreasing stress in time caused by a constant deformation. Also the stress needed to impose a certain strain is a function of the speed with which the strain is imposed. Faster straining requires higher stresses.

The time dependent mechanical behaviour of the material is called viscoelastic behaviour if unloading to zero stress level results in vanishing strains. The memory of the stress history is fading. In case of a nonvanishing strain the unrecoverable part of the strain is called plastic strain. If this plastic strain increases during creep then the term viscoplasticity is used. Part of the memory of the stress history is not fading. However this permanent strain often disappears when the material is heated to elevated temperatures.

During the last 70 years much research has been aimed at describing the viscoelastic behaviour of materials. Particularly the phenomenon of creep has long been subjected to thorough investigations because of its practical implications. And from the early stages it was realized that the description of viscoelastic behaviour in plastics was anything but simple.

In this chapter a literature survey of time dependent behaviour of polymers is given. It starts with linear viscoelasticity, after which the most successful theories to account for the nonlinear behaviour are examined. The theories to describe the effects of temperature and ageing on the viscoelastic behaviour are also given. Finally the numerical treatment of these models is derived. The numerical methods enable the models to be applied for engineering purposes such as FEM analyses.
2.2 Constitutive models

A qualitative picture of a creep curve is given in figure 2.1. The curve starts with the primary stage which distinguishes itself by a convex shape corresponding to a decreasing creep strain rate. Most thermoplastics will ultimately show a transition from convex to concave. At higher stress levels this transition is reached in less time. At the transition the strain rate is more or less constant and this stage is called the secondary creep stage. Finally the tertiary stage is entered with an increasing strain rate which will eventually lead to failure.

![Diagram of creep curve with stages labeled](image)

**Figure 2.1** General creep path.

The total creep strain can be considered as the sum of the elastic strain, the viscoelastic strain and the viscoplastic strain:

\[ \varepsilon_c(t) = \varepsilon_0 + \varepsilon_v(t) + \varepsilon_p(t) \]  

(2.1)

In case of loading and unloading figure 2.2 is obtained. The elastic strain will recover immediately followed by the recovery of the viscoelastic strain. The viscoplastic strain will not recover and cause a permanent deformation. However, because of very slow recovery the distinction between viscoelasticity and viscoplasticity can be difficult.

The first step in the description of viscoelastic behaviour is the description of the creep curve. Findley et al. [2.1] used the power law for various thermoplastics:

\[ \varepsilon_c(t) = \varepsilon_0 + \varepsilon_a t^n \]  

(2.2)

Here \( \varepsilon_0 \) denotes the instantaneous strain and \( \varepsilon_a t^n \) the time dependent creep strain, also called the transient creep strain, with \( n < 1 \). This formula however is mostly not valid for
many time decades. The KWW equation [2.2] can in general describe creep curves over longer time ranges, partly because it contains one more fitting parameter:

$$\varepsilon_c(t) = \varepsilon_0 \exp\left(\frac{t}{t_0}\right)^n$$

(2.3)

The advantage of this approach is that a creep curve is described with only three parameters, i.e., $\varepsilon_0$, $t_0$ and $n$.

![Diagram](image)

**Figure 2.2** General creep recovery curve, with instantaneous strain (1), transient strain (2), total creep strain (3) and residual strain (4).

The effect of the stress on the creep strain was empirically fitted with a power law function resulting in the Nutting equation [2.1]:

$$\varepsilon_c(t) = kt^n \sigma^p$$

(2.4)

Findley et al. [2.1] used a hyperbolic sine function for the stress effect:

$$\varepsilon_c(t) = \varepsilon_0 \sinh\left(\frac{\sigma}{\sigma_0}\right) + \varepsilon_3 t^n \sinh\left(\frac{\sigma}{\sigma_3}\right)$$

(2.5)

This hyperbolic time function will also emerge in the Eyring model where it will appear to have a physical background, see section 2.2.3.

These empirical treatments however also have disadvantages. If the creep behaviour of a polymers is to be described over a wide range of stress levels and/or over many time decades then it is likely that empirical equations no longer suffice. Furthermore a computationally effective incremental formulation for the evaluation of the mechanical
response to arbitrary stress or strain histories cannot be derived if these empirical treatments are used. For this the spring dashpot models are needed, see section 2.3.

2.2.1 Linear viscoelastic behaviour

Superposition principle
For linear viscoelastic behaviour the following three restrictions must hold: The first restriction is that the response to any stress history is independent of the moment of application, i.e., the material iso-age. The second restriction is that the material state does not alter due to an applied stress or strain. As a result the additional strain caused by an additional stress is identical to the strain in case the stress acted solely. The third restriction is the reciprocity; if \( \sigma(t) \) results in \( \varepsilon(t) \) then also \( \varepsilon(t) \) must result in \( \sigma(t) \). A linear viscoelastic material obeys the principle of time-invariance and therefore superposition can be applied, [2.1, 2.3, 2.4].

The creep strain is a linear function of the stress:

\[
\varepsilon_c(t) = D(t)\sigma
\]

(2.6)

Here \( D(t) \) is called the tensile creep compliance function. A linear viscoelastic material obeys the principle of superposition: if at time \( t_1 \) a stress \( \Delta\sigma_1 \) is applied and at time \( t_2 \) an additional stress \( \Delta\sigma_2 \) then the strain response is the sum of the individual responses. In formula:

\[
\varepsilon(t) = D(t-t_1)\Delta\sigma_1 + D(t-t_2)\Delta\sigma_2
\]

(2.7)

In general the strain due to a stepwise stress history is obtained by summation of all individual steps responses. An arbitrary stress history can be considered as a sequence of infinitesimal steps. The summation sign then changes into an integral:

\[
\varepsilon(t) = \int_0^t D(t-\xi)\frac{d\sigma}{d\xi}d\xi
\]

(2.8)

The integral is referred to as the Boltzmann superposition or Boltzmann convolution integral.

The linear viscoelastic model of equation (2.8) is a creep model since the strain is given as a function of the stress. Likewise it is possible to obtain a relaxation model with the stress in terms of the strain:
\[ \sigma(t) = \int_{0}^{t} E(t-\xi) \frac{dE}{d\xi} d\xi \]  \hspace{1cm} (2.9)

Here the \( E(t) \) denotes the stress relaxation function. Both the \( D(t) \) in (2.8) and the \( E(t) \) in (2.9) are called kernel functions. The creep and the relaxation model must be identical when applied on the same material. By substitution of the strain evaluated with (2.8) into (2.9) the original stress must be regained. Using Laplace transform the reciprocity leads to:

\[ \int_{0}^{t} D(\xi) E(t-\xi) d\xi = \int_{0}^{t} D(t-\xi) E(\xi) d\xi = t \]  \hspace{1cm} (2.10)

However conversion of \( D(t) \) into \( E(t) \) can in general not be accomplished analytically and therefore numerical methods must be employed.

**Spring dashpot models**

An appropriate method for engineering purposes to describe \( D(t) \) or \( E(t) \) is by means of the spring dashpot models. The spring and dashpot, see figures 2.3 and 2.4, obey the following equations respectively:

\[ \sigma = E \dot{\varepsilon} \] \hspace{1cm} (2.11)

\[ \sigma = \eta \frac{d\varepsilon}{dt} \]

These two elements can be placed in parallel to form a Kelvin-Voigt element or in series to form a Maxwell element [2.1, 2.3, 2.4], see figures 2.5 and 2.6. In case of creep the Kelvin-Voigt element gives a viscoelastic response whereas the Maxwell element results in an elastic and viscoplastic response. The Maxwell element exhibits stress relaxation when given a constant strain. The combination of a Kelvin-Voigt element in series with a Maxwell element is called the Burger's model.

\[ \begin{array}{c}
\text{Spring element.} \\
\end{array} \quad \begin{array}{c}
\text{Dashpot element.} \\
\end{array} \]
Since both the spring and the dashpot are described by a linear equation the Kelvin-Voigt and the Maxwell element are also linear and obey the superposition principle. A series of Kelvin-Voigt elements placed in series together with a single spring and a single dashpot yields the Generalized Kelvin-Voigt model, see figure 2.7. Figure 2.8 shows the Generalized Maxwell model.

![Kelvin-Voigt element](image1.png) ![Maxwell element](image2.png)

**Figure 2.5** *Kelvin-Voigt element.*  **Figure 2.6** *Maxwell element.*

The creep compliance function of the Generalized Kelvin-Voigt model and the relaxation modulus function of the Generalized Maxwell model are then given by a series expansion:

\[
D(t) = D_0 + \sum_k D_k \left( 1 - \exp \left( -\frac{t}{\tau_k} \right) \right) + \frac{t}{\eta} \quad (2.12)
\]

\[
E(t) = \sum_l E_l \exp \left( -\frac{t}{\tau_l} \right) \quad (2.13)
\]

with:

\[
D_k = \frac{V}{E_k} \quad \tau = \frac{\eta}{E} \quad (2.14)
\]

The parameter \( \tau \) is the characteristic time of the element; in case of a Kelvin-Voigt element it is also called retardation time and in case of a Maxwell element it is also called relaxation time. The series of equation (2.12) is called the Prony series. The Generalized Kelvin-Voigt model has a strict separation between elasticity, viscoelasticity and viscoplasticity whereas these contributions are interwoven in case of the Generalized Maxwell model. The numerical treatment of the spring-dashpot models, which is used later in FEM calculations, is given in section 2.3.
In figure 2.9 the creep compliance curve of HDPE measured at 4 MPa is shown together with its Generalized Kelvin-Voigt fit. The creep compliance curve is obtained by the summation of all individual contributions, which are shown below the measurements as dashed lines. The characteristic times $\tau_k$ are chosen equidistantal on logarithmic time scale while the contributions of the elements $D_k$ as well as the elastic contribution $D_0$ are used to fit the experimental data. The $D_k$ are plotted as diamonds as a function of $\tau_k$ resulting in the creep spectrum. The complex shape of the creep curve of figure 2.9 can be fitted accurately. Possible nonlinear effects of stress/strain are discussed in the next section.
2.2.2 Nonlinear viscoelastic behaviour

Since many polymers exhibit nonlinear viscoelastic behaviour, the Boltzmann integral does not suffice to describe these polymers. Much research has been addressed to develop models for nonlinear viscoelasticity, which can describe the nonlineairities found during measurements on polymers. Generally on starts with the Boltzmann integral and nonlinearity enters into the model by additional stress or strain functions. The more functions added, the more nonlinear phenomena can be described, but on the other hand the more complex the model becomes. The most successful nonlinear models are discussed in this section.

Multiple integral model

One of the first models developed for nonlinear viscoelasticity is the multiple integral model [2.5], which can be considered as a series expansion of the Boltzmann integral [2.1]:

Figure 2.9  Creep strain divided by the stress measured during creep of HDPE at 4 MPa (o), the Kelvin-Voigt fit (solid line), the individual contributions (dashed lines) and the creep spectrum $D_k$ (●) as a function of $\tau_k$. 

\[
\varepsilon(t) = \int_0^t \varphi_1(t - \xi) \frac{d\sigma}{d\xi} d\xi + \int_0^t \varphi_2(t - \xi, t - \xi') \frac{d\sigma}{d\xi} \frac{d\sigma}{d\xi'} d\xi d\xi' + \\
+ \int_0^t \int_0^t \varphi_3(t - \xi, t - \xi_2, t - \xi_3) \frac{d\sigma}{d\xi} \frac{d\sigma}{d\xi_2} \frac{d\sigma}{d\xi_3} d\xi d\xi_2 d\xi_3 + \ldots
\]

(2.15)

This model can describe any nonlinear viscoelastic iso-age material. The first term in equation (2.15) is the Boltzmann superposition integral. The creep strain of this model is given by:

\[
\varepsilon_c(t) = \sigma \varphi_1(t) + \sigma^2 \varphi_2(t, t) + \sigma^3 \varphi_3(t, t, t) + \ldots
\]

(2.16)

Any arbitrary set of creep curves can be described by such a series expansion.

The generality seems to be an advantage, but it is also the major drawback. Even when only three terms are taken into account, a very large number of experiments is needed to completely determine all kernel functions. This is illustrated for a two-step stress history with a stress \( \Delta\sigma_0 \) applied at zero time increased with \( \Delta\sigma_1 \) at \( t_1 \):

\[
\varepsilon(t) = \Delta\sigma_0 \varphi_1(t) + \Delta\sigma_1 \varphi_1(t - t_1) + \\
+ \Delta\sigma_0^2 \varphi_2(t, t) + 2\Delta\sigma_0 \Delta\sigma_1 \varphi_2(t, t - t_1) + \Delta\sigma_1^2 \varphi_2(t - t_1, t - t_1) + \ldots
\]

(2.17)

And here only the single and double integral are considered. The time function is assumed symmetric with respect to its time arguments: \( \varphi_2(t, t - t_1) = \varphi_2(t - t_1, t) \). Moreover the evaluation of the higher order integrals is time consuming and hence for engineering purposes the multiple integral is impracticable.

**Leaderman model**

Leaderman found, based on his tests on plasticized PVC, see [2.6, 2.7], that the nonlinearity of viscoelastic behaviour in case of creep can be modelled by a single integral equation. He stated that a nonlinear strain measure obeys the linear Boltzmann equation [2.4]:

\[
H(\varepsilon(t)) = \int_0^t D(t - \xi) \frac{d\sigma}{d\xi} d\xi
\]

(2.18)

This model can be inverted to:

\[
\sigma(t) = \int_0^t E(t - \xi) \frac{dH(\varepsilon)}{d\xi} d\xi
\]

(2.19)
Likewise a nonlinear stress measure can be applied [2.4]:

$$
\varepsilon(t) = \int_0^t D(t-\xi) \frac{dG(\sigma)}{d\xi} \, d\xi
$$

(2.20)

Substitution of the relaxation conditions into (2.19) yields:

$$
\sigma_r(t) = E(t) H(\varepsilon)
$$

(2.21)

Substitution of the creep conditions into (2.20) yields:

$$
\varepsilon_c(t) = D(t) G(\sigma)
$$

(2.22)

which in fact is nothing more than separation of variables with the linear stress/strain replaced by a nonlinear stress/strain function. For a two step stress history the response of the Leaderman creep model can be obtained by replacing $\Delta \sigma$ with $\Delta G$ in equation (2.7).

The Leaderman creep model can be rewritten by replacing $G(\sigma)$ with $\sigma g(\sigma)$ in equation (2.20). By restricting $g(\sigma)$ with $g(0) = 1$, the equation reduces to the linear case at low stresses. The creep strain divided by the creep stress of a material obeying the creep form of the Leaderman model is shown in figure 2.10. The curves can be shifted vertically on logarithmic strain scale with $\Delta g = g(\sigma_2) - g(\sigma_1)$ upon a single curve: the master curve. Only creep curves are needed to determine all material parameters.

The Leaderman model has been applied successfully on HDPE. Zapas and Crissman [2.8] could describe creep curves of HDPE by vertical shifting of the (transient) creep compliance, in combination with nonlinear elasticity. For the description of recovery an additional term for viscoplasticity was needed. Stress relaxation curves of HDPE could also be superimposed by vertical shifting on logarithmic stress scale as was shown by Popelar et al. [2.9].

![Figure 2.10](image)

**Figure 2.10**  Effect of the Leaderman function $g(\sigma)$ on creep.
2.2 Constitutive models

Modified Superposition Model
This model places no restrictions on the creep strain [2.1, 2.10]:

$$\varepsilon_c(t) = G(\sigma, t)$$  \hspace{1cm} (2.23)

This function can be written in terms of a series like in equation (2.16) of the multiple integral model. However the number of material functions in the multiple integral model is decreased by assuming that a stepwise stress change can be considered as the simultaneous removal of the "old" stress and application of the "new" stress:

$$\varepsilon(t) = G(\sigma_0, t) - G(\sigma_0, t - t_i) + G(\sigma_1, t - t_i)$$  \hspace{1cm} (2.24)

This equation gives the strain if $\sigma_0$ applied at zero time is changed into $\sigma_1$ at time $t_i$. For an arbitrary stress path the assumption leads to:

$$\varepsilon(t) = \int_0^t \frac{\partial G(\sigma, t - \xi)}{\partial \sigma} \frac{d\sigma}{d\xi} d\xi$$  \hspace{1cm} (2.25)

If $G(\sigma, t) = D(t) G(\sigma)$ is substituted into (2.25) then the Leaderman model, (2.20), is regained. Since any function $G(\sigma, t)$ can be written as a series expansion:

$$G(\sigma, t) = \sum_i D_i(t) G_i(\sigma)$$  \hspace{1cm} (2.26)

the modified superposition model is nothing more than a series of Leaderman models. Therefore this model needs creep curves only to determine all material parameters. In principle, like the multiple integral model, any arbitrary set of creep curves can be described.

Schapery creep model
On the basis of thermodynamic considerations Schapery introduced his model for nonlinear viscoelasticity [2.4, 2.11 - 2.13]:

$$\varepsilon(t) = D_0 \sigma_0(\sigma) + g_1(\sigma) \int_0^t \Delta D(\psi(t) - \psi(\xi)) \frac{d(\sigma_2(\sigma))}{d\xi} d\xi$$  \hspace{1cm} (2.27)

with the so called reduced times:

$$\psi(t) = \int_0^t \frac{d\xi}{a_0(\sigma)}$$  \hspace{1cm} $$\psi(\xi) = \int_0^\xi \frac{d\xi}{a_0(\sigma)}$$  \hspace{1cm} (2.28)
In this model the strain is divided into an elastic contribution, the first term behind the equal sign, and a time dependent contribution, the integral term. The nonlinearity is described with four functions: \( g_0(\sigma) \), \( g_1(\sigma) \), \( g_2(\sigma) \) and \( a_\sigma(\sigma) \). To obtain the Boltzmann integral at vanishing stresses all four functions must equal one at zero stress level. When the stress and strain variables in (2.27) and (2.28) are interchanged then the relaxation form of the Schapery model is obtained, see also section 2.3.1.

The Schapery model yields the following creep strain:

\[
\varepsilon_c(t) = D_0 \varepsilon_0(\sigma) + \Delta D \left( \frac{t}{\alpha(\sigma)} \right) \varepsilon_1(\sigma) g_3(\sigma)
\]

(2.29)

Note that \( g_1(\sigma) \) and \( g_3(\sigma) \) have the same effect on creep. Therefore on based on creep alone no distinction between \( g_1(\sigma) \) and \( g_3(\sigma) \) can be made. If \( \sigma_0 \), applied at zero time, is changed into \( \sigma_t \) at time \( t_1 \) then the Schapery model gives the following strain:

\[
\varepsilon(t) = D_0 \varepsilon_0 g_0^1 + g_1^0 \left[ \Delta D \left( \frac{t_1}{a_\sigma} + \frac{t-t_1}{a_\sigma} \right) \sigma_0 g_2^0 + \Delta D \left( \frac{t-t_1}{a_\sigma} \right) (\sigma_1 g_2^1 - \sigma_0 g_2^0) \right]
\]

(2.30)

where the superscript of the four stress functions denotes the stress level to be substituted into the function.

The function \( a_\sigma(\sigma) \) is the time-stress superposition function which represents the principle of the intrinsic time clock, see also [2.14]. The nonlinear effect of a stress increase on the creep curve is a decrease of the time scale; all time dependent processes are accelerated by the stress. When only the time-stress superposition is implemented in the Boltzmann integral to describe nonlinear viscoelasticity, the model is also called the Leonov model [2.15]. The effect is visualized in figure 2.11.

**Figure 2.11** Effect of the time-stress superposition function \( a_\sigma(\sigma) \) on creep.
The function $g_0(\sigma)$ can account for nonlinear elasticity. Also if part of the "instantaneous" (first measured) creep strain in fact contains a viscoelastic part taking place at a very small time lapse, the nonlinearity caused by $g_1(\sigma)$ and/or $g_2(\sigma)$ can be accounted for by this stress function.

The function $g_1(\sigma)$ is the same function as $g(\sigma)$ in the Leaderman model but in the Schapery model it only acts on the viscoelastic strain. Since $g_1(\sigma)$ and $g_2(\sigma)$ have the same effect on creep, the effect of the Leaderman function is considered for creep recovery in figure 2.12. The dashed line is obtained if the Leaderman function would also apply to the elastic strain. Both the creep strain and the recovery strain are increased by a factor $\Delta g_2$.

The pre-integral function $g_i(\sigma)$ on the other hand only applies to the creep strain. Therefore when two step stress history experiments are performed then a distinction between $g_1(\sigma)$ and $g_2(\sigma)$ can be made. The pre-integral function can account for the effect that the instantaneous recovery is larger than the instantaneous creep [2.3, 2.16], see figure 2.13.

![Figure 2.12](image1.png)  
**Figure 2.12**  Effect of the Leaderman (Schapery integral) function $g_2(\sigma)$ on creep recovery.

![Figure 2.13](image2.png)  
**Figure 2.13**  Effect of the Schapery pre-integral function $g_1(\sigma)$ on creep recovery.
Lai et al. [2.17] fitted creep curves of HDPE by vertical and horizontal shifting of one single master curve. Creep curves of PE, measured over a long period of time, exhibited vertical shifts, see Turner [2.18]. Creep curves of PP shifted vertically and horizontally, see [2.16]. The description of creep recovery of HDPE by Lai et al. [2.19] required the addition of a viscoplastic term to the Schapery creep model. PC creep curves can be described by horizontal shifting only, i.e., Leonov model, as was shown by Tervoort et al. [2.20].

**BKZ theory**

Two models were derived by Bernstein, Kearsley and Zapas [2.4, 2.21]: a BKZ solid model and a BKZ liquid model. Both models are relaxation form models directly derived for multiaxial stress strain situations. In both models it is assumed that the material is incompressible and initially isotropic.

The BKZ solid model is derived from the relaxation form of the multiple integral model. Only the single integral term is taken. As a result the model is fully determined by three material constants and two kernel functions. The nonlinearity in the model is caused by the Green-St. Venant tensor which is used as strain measure. No strain functions for fitting, like in the relaxation form of the Schapery model, are present.

In case of stress relaxation the three-dimensional strain situation is known since the material has a constant volume and is isotropic. The true stress takes the following form:

$$\frac{\sigma(t)}{\lambda^2 - 1/\lambda} = (\lambda^2 - 1) \left[ \frac{1}{2} A(t) + B(t) \right] + \frac{1}{\lambda} [A(t) + B(t)] + M - A(t)$$  \hspace{1cm} (2.31)

with $\lambda = 1 + e$ the extension and $A(t)$ and $B(t)$ time functions which can be used to fit relaxation data. If $\frac{1}{2} A(t) + B(t) = 0$ then the material, in that situation, will seem a Mooney material whereas it will seem Neohookian if $A(t) = B(t) = 0$. Both the Mooney [2.22] and the Neohookian [2.22] material models are used to describe the elastic behaviour of rubbers. As a consequence the BKZ solid model is generally used to describe viscoelasticity in rubbers with large strain levels.

The BKZ liquid model starts with the constitutive equation for finite linear viscoelasticity derived by Coleman and Noll [2.23] for isotropic materials with fading memory. It is assumed that the effect of the configuration (strain) at time $\xi < t$ on the present stress is equivalent to the effect of stored elastic energy with the configuration at time $\xi$ as the preferred configuration. Furthermore the total history contribution equals the sum of all individual contributions, i.e., the superposition principle is applicable. Furthermore constant volume is assumed.
2.2 Constitutive models

In case of stress relaxation the following true stress is obtained:

$$\frac{\sigma(t)}{\lambda^2 - \frac{1}{\lambda}} = (\lambda^2 - 1)[\frac{1}{2} A(t) + B(t)] + \frac{Y}{\lambda} [A(t) + B(t)] + M(t) - A(t)$$  \hspace{1cm} (2.32)

As can be observed in case of stress relaxation the BKZ solid and liquid models are identical except for $M(t)$ which in the solid theory is a constant $M$. Therefore the liquid model has also been used to describe solids.

The BKZ model was applied successfully on strain step data of plasticized PVC [2.24]. Zapas [2.25] later modified the model by incorporating the time-strain superposition principle. This principle is also present in the relaxation form of the Schapery model.

Lianis model

This model is similar to the BKZ liquid model. Again one starts with the constitutive equation for finite linear viscoelasticity [2.23]. Lianis [2.4, 2.26-2.28] assumed that the material is incompressible, that the stress relaxation functions are independent of the strain invariants and that the short and long term behaviour is Neohookian. In case of stress relaxation the same as the BKZ liquid model was found. Later the model was modified by choosing a different long term behaviour [2.29].

2.2.3 Yielding

The yield point in case of polymers is (mostly) defined as the point in the stress-strain curve where the load reaches its maximum, [2.3]. The yield point therefore has a maximum resistance to a constant strain rate. During creep the strain rate is minimal for the secondary creep stage, see figure 2.1, leading to a maximum resistance against a constant load and therefore this situation can also be called yielding.

The yield behaviour of many polymers can be described by the Eyring model [2.3, 2.30]. This model starts with the assumption that a local molecular motion occurs when a certain amount of energy is surpassed. The frequency $\nu$ of exceeding this energy barrier $\Delta H$ at a certain temperature $T$ is given by the Arrhenius equation [2.3]:

$$\nu = \nu_0 \exp\left(\frac{-\Delta H}{RT}\right)$$  \hspace{1cm} (2.33)

The next assumption concerns the occurrence of a stress $\sigma$. The energy barrier for the motion in the stress direction is decreased with $\lambda\sigma$ whereas the barrier in the other
direction is increased with \( \lambda \sigma \). Assuming that a molecular motion induces a strain, the net result of the molecular motions yields a strain rate:

\[
\dot{\varepsilon} = \dot{\varepsilon}_0 \exp\left(-\frac{\Delta H}{RT}\right) \sinh\left(\frac{\lambda \sigma}{RT}\right)
\]

(2.34)

which is the Eyring equation.

The Eyring model (for constant temperature) can be considered as a single dashpot obeying the time-stress superposition principle. This dashpot therefore is also called an Eyring dashpot:

\[
\dot{\varepsilon} = \frac{\sigma}{\eta'_{\sigma}(\sigma)} = \frac{\sigma}{\eta_{\sigma_e}(\sigma)} = \frac{\sigma \sinh(\sigma/\sigma_0)}{\eta_0 \sigma/\sigma_0}
\]

(2.35)

with \( a_e(\sigma) \) the time-stress superposition function defined in the Schapery creep model.

A graph with the yield strain rate on logarithmic scale against the yield stress on linear scale (Eyring plot, see figure 4.10) shows a straight line when the yield behaviour obeys the Eyring equation. PC is an example of such a material [2.31]. The \( \sigma_0 \) is often so small that the hyperbolic sine can be approximated by the exponential function.

However, for many polymers two Eyring processes are necessary to describe the yield behaviour. This is found for PVC [2.31], for ultra high modulus PE fibers [2.32] and other thermoplastics [2.33 - 2.35]. The yield points in an Eyring plot then exhibit a straight line with a bend in it, see figure 4.10. This behaviour can be described by two Eyring dashpots in parallel. Apparently two molecular processes are then responsible for the mechanical behaviour.

### 2.2.4 The effect of temperature

A temperature increase not only induces a volumetric expansion but it also increases the creep strain rate. The effect of temperature on viscoelasticity was described by Leaderman who first introduced the principle of time-temperature superposition [2.2]:

\[
\varepsilon(t) = \int_0^t D(\psi(t) - \psi(\xi)) \frac{d\sigma}{d\xi} d\xi
\]

(2.36)

with the term reduced time:
\[ \psi(t) = \int_0^t \frac{d\xi}{a_r(T)} \]
\[ \psi(\xi) = \int_0^\xi \frac{d\xi}{a_r(T)} \]

(2.37)

This principle means that a creep curve at a higher temperature shifts horizontally on logarithmic time scale. However measurements show that small vertical shift on logarithmic strain scale [2.36] occur as well:

\[ \varepsilon(t) = \int_0^t D(\psi(t) - \psi(\xi)) \frac{d(\sigma_{e_T}(T))}{d\xi} \, d\xi \]

(2.38)

Since sudden temperature quenches during creep hardly result in sudden steps in the (mechanical) strain [2.37], placing \( g_r(T) \) in the integral is more obvious than placing it in front of the integral (compare \( g_t(\sigma) \) and \( g_s(\sigma) \) in the Schapery creep model).

In case of an Eyring process the time-temperature function can be derived by using equation (2.33), resulting in the Arrhenius time-temperature superposition formula:

\[ \log(a_r) = \log \left( \frac{\nu(T_0)}{\nu(T)} \right) = \log \left( \frac{\exp \left( \frac{-\Delta H}{RT_0} \right)}{\exp \left( \frac{-\Delta H}{RT} \right)} \right) = \frac{\Delta H}{R} \left( \frac{1}{T} - \frac{1}{T_0} \right) \]

(2.39)

where \( T_0 \) is the reference temperature. The horizontal shift values of creep curves at different temperatures plotted on logarithmic scale against the reciprocal temperature (Arrhenius plot, see figure 4.55) will be situated on a straight line when an Eyring process is responsible for the strain.

Another often applied time-temperature superposition function is the WLF equation [2.38], which is based on free volume considerations. The free volume is “the volume not occupied by molecules”. Above the glass transition temperature \( T_g \) this free volume enables the movement of large molecular segments and the material is liquid. The material is in thermodynamic equilibrium. During cooling this free volume decreases until at \( T_g \) it becomes too small to make large molecular motions possible and the material as well as a small part of the free volume itself is frozen in. The material is no longer in thermodynamic equilibrium.

Above the glass transition the free volume varies linearly with the temperature [2.3]:

\[ f = f_g + (T - T_g)\alpha_f \]

(2.40)

with \( f = V_f/V \) the free volume fraction. Doolittle [2.3] showed that the flow viscosity of amorphous polymers is directly related to the free volume:
\[
\ln \eta = \ln A + B \left( \frac{V - V_f}{V_f} \right) = \ln A + B \left( \frac{1 - f}{f} \right)
\]  
\tag{2.41}

Substitution yields:

\[
\log(a_T) = \log \left( \frac{\eta(T)}{\eta(T_0)} \right) = \frac{B}{2.303} \left( \frac{1}{f} - \frac{1}{f_0} \right) = \frac{-C_1(T - T_0)}{C_2 + (T - T_0)}
\]  
\tag{2.42}

with again \( T_0 \) the reference temperature for which often \( T_g \) is taken. The WLF equation is valid for motions which are related to the glass transition. Furthermore it is valid above \( T_g \) and to some extent below it.

If the temperature effect can be described by simple horizontal and vertical shifting of a master curve then the material is defined as thermo-rheologically simple, [2.39]. If the deformation process is governed by two or more processes each having their own time-stress superposition functions, then no master curve can be constructed and the material is thermo-rheologically complex. Time-temperature superposition is believed not to hold for semi-crystalline polymers like HDPE, [2.40].

### 2.2.5 Ageing

Cooling of an amorphous polymer below its glass transition temperature solidifies it due to lack of free volume which is necessary for the mobility of large molecular segments. However some mobility is still present, but with a very large characteristic time. Kovacs [2.41] showed that below the glass transition the free volume still decreases slowly in time, seeking to reach its equilibrium value. As a consequence the time scale of molecular motions increases in time, resulting in stiffer behaviour. This phenomenon is called physical ageing. The ageing time \( t_a \) is defined as the time from the moment that the material was last quenched through \( T_g \). Heating above \( T_g \) will erase the previous ageing.

It was Struik [2.42] who first systematically examined the effect of physical ageing on the creep behaviour of polymers. To describe this effect he introduced time-ageing time superposition, which is similar to time-temperature superposition:

\[
\varepsilon(t) = \int_0^t D(\psi(t) - \psi(\xi)) \frac{d\sigma}{d\xi} d\xi
\]  
\tag{2.43}

with again the term reduced time:
\[
\psi(t) = \int_0^t \frac{d\xi}{a_e(\xi)} \\
\psi(\xi) = \int_0^\xi \frac{d\xi}{a_e(\xi)}
\]

(2.44)

By shifting creep curves horizontally on logarithmic time scale it was often possible to construct a master curve. These creep curves must have a creep time substantially smaller, \( t_c \leq 0.3t_e \), than the ageing time so that during creep the effect of ageing can be neglected (iso-age). Depending on the type of material a small vertical shift on logarithmic strain scale or on linear strain scale was necessary [2.42 - 2.46].

For many polymers \( a_e \) vs. \( t_e \) showed straight lines on double logarithmic scale. Struik therefore introduced the ageing rate [2.42]:

\[
\mu = \frac{d \log(a_e)}{d \log(t_e)}
\]

(2.45)

At low stress levels the ageing rate turns out to be independent of the stress level of the creep curves. At higher stress levels the induced deformations can cause a small increase in the free volume resulting in de-ageing also called rejuvenation [2.47 - 2.49].

Although the glass transition temperature \( T_g \) of HDPE is situated at approximately -115\(^\circ\)C, this material exhibits physical ageing at room temperature. This is also the case for other semi-crystalline materials. Struik [2.43 - 2.46] supposed that the crystalline phase reduces the mobility of the amorphous regions next to the crystals and causes an increase of the \( T_g \) in these disturbed amorphous regions. As a consequence a glass transition temperature region is present \( \{ T_{gL}, \ldots, T_{gU} \} \) and room temperature can be within this temperature interval. The ageing behaviour becomes complex with ageing rates \( \mu \) depending on the temperature and vertical shift values changing sign.

Some other scientists objected to the ageing theory of Struik. The horizontal shift values were considered to be too large from a physical point of view [2.50]. Other theories were launched like the sequential ageing theory [2.50, 2.51] which states that molecular motions with a small retardation time age faster that those with large retardation times. Also the creep curve could change shape due to ageing [2.51, 2.52]. Struik on his turn objected to the sequential ageing theory [2.53]. Struik’s theory is still the most applied model to describe ageing because of its simplicity and ability to give good predictions for long term creep with ageing during creep.

The physical ageing of HDPE was described by Struik [2.43, 2.45] and Lai et al. [2.17] with time-ageing time superposition and vertical shifting on linear strain scale. Lai found that at low stress levels the ageing rate of HDPE is independent of the stress level.
2.2.6 Multiaxial stress states

Experiments with homogeneous multiaxial stresses and strains are hard to carry out and literature on viscoelastic behaviour during three-dimensional stress situations is therefore scarce. Stress situations other than uniaxial tension are mostly accomplished by torsion of hollow cylinders. Furthermore different hydrostatic pressures during tensile straining can be applied. The scarce literature on time dependent mechanical behaviour with multiaxial stresses or strains is discussed. After the concept of linear viscoelasticity is extended for multiaxial stress states, the nonlinear effects in three-dimensional stress strain situations are discussed.

Linear viscoelastic behaviour

The behaviour of an isotropic linear elastic material is fully determined by two material parameters [2.1, 2.4]:

\[ s_y = 2G_o d_y \]
\[ \sigma_m = 3K_o \varepsilon_m \]

(2.46a)

(2.46b)

with \( s_y \) is the deviatoric stress, \( \sigma_m \) the hydrostatic stress (minus pressure \( p \)), \( d_y \) the deviatoric strain and \( \varepsilon_m \) the volumetric strain and with the material parameters \( G_o \) the shear modulus and \( K_o \) the bulk modulus. In this approach the stress and strain tensors \( \sigma_y \) and \( \varepsilon_y \) are split into their volumetric and deviatoric part:

\[ \varepsilon_y = d_y + \delta_y \varepsilon_m \]
\[ \sigma_y = s_y + \delta_y \sigma_m \]

(2.47a)

(2.47b)

The strains can also be written in terms of the stresses:

\[ d_y = \frac{1}{2} J_o s_y \]
\[ \varepsilon_m = \frac{1}{3} B_o \sigma_m \]

(2.48a)

(2.48b)

with the material parameters \( J_o \) the shear compliance and \( B_o \) the bulk compliance. Moduli and compliances are interrelated:

\[ J_o = \frac{1}{G_o} \]
\[ B_o = \frac{1}{K_o} \]

(2.49a)

(2.49b)
In case of an isotropic linear viscoelastic material the linear terms can be replaced by convolution integrals [2.1, 2.4]:

\[
d_y(t) = \frac{1}{2} \int_0^t J(t - \xi) \frac{ds_y}{d\xi} d\xi
\]  \hspace{1cm} (2.50a)

\[
\varepsilon_m(t) = \frac{1}{2} \int_0^t B(t - \xi) \frac{d\sigma_m}{d\xi} d\xi
\]  \hspace{1cm} (2.50b)

The relaxation model can be formed likewise. The reciprocity equation (2.10) forms the link between the creep and relaxation models.

**Nonlinear viscoelastic behaviour**

The creep strain of a nonlinear viscoelastic solid during a multiaxial stress can be written in the following general expression [2.47, 2.54] (for small strain levels):

\[
\varepsilon_y(t) = \frac{1}{2} J(t, I_1, I_2, I_3) s_y + \frac{1}{2} B(t, I_1, I_2, I_3) \sigma_m \delta_y
\]  \hspace{1cm} (2.51)

With the stress invariants [2.55]:

\[
I_1 = \text{tr}(\sigma) = \sigma_{ii} = \sigma_1 + \sigma_2 + \sigma_3
\]  \hspace{1cm} (2.52a)

\[
I_2 = \frac{1}{2} \text{tr}(\sigma \sigma) - \frac{1}{2} I_1^2 = \frac{1}{2} \sigma_y \sigma_y - \frac{1}{2} I_1^2 = \sigma_1 \sigma_2 + \sigma_2 \sigma_3 + \sigma_3 \sigma_1
\]  \hspace{1cm} (2.52b)

\[
I_3 = \text{det}(\sigma) = \sigma_1 \sigma_2 \sigma_3
\]  \hspace{1cm} (2.52c)

where \( \sigma_1, \sigma_2 \) and \( \sigma_3 \) are the principle stresses.

Mallon et al. [2.56] found that the bulk creep response of PP and PVC were linear viscoelastic. Furthermore their volume response had a small transient component compared with the instantaneous contribution: \( \varepsilon_{vo} > 7 \Delta \varepsilon_i(t) \) even at creep times of 10^6 s. Benham [2.57] could predict torsion creep using tension and compression creep data. For all experiments they calculated the octahedral stress and strain, which are proportional to the equivalent stress and strain defined as follows:

\[
\tau_{eq} = \sqrt{\frac{1}{3} s_y s_y} = \sqrt{I_2} = \sqrt{\frac{1}{6} \left( (\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2 \right)}
\]  \hspace{1cm} (2.53a)

\[
\gamma_{eq} = \sqrt{2 d_y d_y}
\]  \hspace{1cm} (2.53b)

With \( I_2 \) the second invariant of the deviatoric stress and the equivalent stress equal to the Von Mises stress. It was found that the relation for torsion was equal to the average of the relations for tension and compression. This effect was attributed to the effect of the
hydrostatic pressure $p = -\frac{1}{3} I_1$, which should then have a linear effect on $J(t)$. Usage of $\tau_{eq}$ for the determination of the nonlinearity yielded better results than usage of the maximum shear stress $\tau_{\text{max}}$.

Based on experimental results Buckley et al. [2.54] assumed the following expression for multiaxial creep:

$$\varepsilon(t) = \frac{1}{2} J(t, I_1, I_2) \sigma_{ij} + \frac{1}{2} B(t) \sigma_{ii} \delta_{ij}$$

which is a more specific form of equation (2.53). Pao et al. [2.58] further simplified this expression by assuming that the viscoelastic process is incompressible and independent of the pressure:

$$\varepsilon(t) = \frac{1}{2} J(t, I_2) \sigma_{ij} + \frac{1}{2} B_0 \sigma_{jj} \delta_{ij}$$  \hspace{1cm} (2.55)

If $J(t, I_2)$ is replaced by $J_0 + J_{pl}(I_2)$ then equation (2.55) corresponds to the Lévy-Mises equations for plasticity/yielding [2.3]. $J_{pl}(I_2)$ is the term for the plastic strain.

Lateral contraction measurements during creep of HDPE performed by Lai et al. [2.59] yielded Poisson’s ratio values of approximately 0.45. This indicates that for HDPE the volume response is much stiffer than the shear response.

Plasticity/yielding

The Lévy-Mises model for plasticity/yielding assumes that plasticity occurs when the equivalent stress as defined in (2.55a) surpasses a critical value:

$$\tau_{eq} > \tau_0$$  \hspace{1cm} (2.56)

The equivalent plastic strain solely depends on the equivalent stress and the plastic strain components are proportional to the deviatoric stresses. In fact this criterion for plasticity is based upon the idea that plasticity occurs when the energy $U'$ caused by the deviatoric stress surpasses a critical value. In case of linear elasticity:

$$U' = \frac{1}{2} \tau_{eq} \gamma_{eq} = \frac{1}{2} G \gamma_{eq}^2 = \frac{1}{2} \frac{\tau_{eq}^2}{G}$$

The Tresca plasticity criterion assumes that plasticity/yielding depends on the maximum shear stress $\tau_{\text{max}}$.

Experimental results with yielding of polymers under multiaxial stresses indicated that the yield stresses obey the Lévy-Mises or Tresca criteria, [2.3]. Measurements on many polymers showed, however, that the effect of the hydrostatic pressure on the yield stress
during tensile straining is often not negligible. The yield stress increases linearly with the hydrostatic pressure \([2.60]\). As a result the Coulomb criterion for plasticity/yielding was proposed \([2.61]\):

\[
\tau_{\text{ext}} > \tau_0 + \alpha p
\]  

(2.58)

The effect of the hydrostatic pressure is then implemented.

As was stated in section 2.2.3 the yield behaviour of polymers can often be described by an Eyring process. The Eyring viscoplasticity model can be extended for three-dimensional stresses including the effect of pressure. In general:

\[
\dot{\varepsilon}_{ij} = \frac{\sigma_{ij}}{\eta_0 a_t(I_1, I_2, I_3)}
\]  

(2.59)

with \(a_t(I_1, I_2, I_3)\) the time-stress superposition function (see section 2.2.2, Schapery creep model). The following equation is found when the energy barrier \(\Delta H\) of equation (2.33) increases linearly with the pressure \(p\) \([2.3]\):

\[
a_t(I_1, I_2, I_3) = a_{eq}(\tau_{eq}) a_p(p) = \frac{\tau_{eq}/\tau_0}{\sinh(\tau_{eq}/\tau_0)} \exp\left(-\frac{p}{p_0}\right)
\]  

(2.60)

Resulting in:

\[
\dot{\gamma}_{eq} = \dot{\gamma}_0 \sinh\left(\frac{\tau_{eq}}{\tau_0}\right) \exp\left(-\frac{p}{p_0}\right)
\]  

(2.61)

The result is a linear relation between the tensile yield stress and the hydrostatic pressure, since for large stresses \(\sinh(x) \approx \frac{1}{2} \exp(x)\).

Mears et al. \([2.60]\) measured a linear increase of the tensile yield stress of PE and PP with the hydrostatic pressure.

### 2.3 Numerical modelling

Since the Schapery model is the most general model and includes several other models, incremental formulations are derived for this model using the differential form and the integral form. An incremental formulation of the convolution integral of equation (2.27) is computationally much more effective than straightforward numerical integration, since numerical integration requires the computation of the complete integral for each time value for which the material response is wanted.
2.3.1 Schapery model, integral form

Creep formulation
For an incremental formulation the Generalized Kelvin-Voigt model is needed for the kernel function. The Schapery model, see section 2.2.2, can be written as:

\[ \varepsilon(t) = D_0 \sigma g_0(\sigma) + g_1(\sigma) \sum_k q_k(t) \]  

(2.62)

with for the k-th Kelvin-Voigt element the nonlinear convolution integral:

\[ q_k(t) = D_k \int_0^t \left( 1 - \exp \left( - \frac{\psi(t) - \psi(\xi)}{\tau_k} \right) \right) \frac{dG_2(\sigma)}{d\xi} d\xi \]  

(2.63)

For convenience:

\[ G_2(\sigma) = \sigma g_2(\sigma) \]  

(2.64)

The reduced time \( \psi \) was defined in equation (2.28). The following numerical scheme was first applied by Henriksen [2.62] and later by Lai et al. [2.63] and is presented here with only few algebraic rearrangements.

The subscript \( n \) denotes the point of time number:

\[ q_k^{n+1} = \int_D \left( 1 - \exp \left( - \frac{\psi(t_{n+1}) - \psi(\xi)}{\tau_k} \right) \right) \frac{dG_2(\sigma)}{d\xi} d\xi \]  

(2.65)

Partial integration, splitting the integral and backsubstitution of \( q_k^n \) results in the following recurrent expression:

\[ q_k^{n+1} = \exp \left( - \frac{\Delta \psi_n}{\tau_k} \right) q_k^n + D_k \int_{t_n}^{t_{n+1}} \exp \left( - \frac{\psi(t_{n+1}) - \psi(\xi)}{\tau_k} \right) \frac{1}{\tau_k a_o(\sigma)} G_2(\sigma) d\xi \]  

(2.66)

with \( \Delta \psi_n = \psi_{n+1} - \psi_n \). The integral in equation (2.66) can be evaluated numerically. A forward integration step is performed by assuming that \( G_2(\sigma) = G_2^* \) and \( a_o(\sigma) = a^* \). To minimize numerical round off errors the result can best be expressed as:

\[ q_k^{n+1} = q_k^n + (D_k G_2^* - q_k^n) \left( 1 - \exp \left( - \frac{\Delta \psi_n}{\tau_k a^*} \right) \right) \]  

(2.67)
with $\Delta t_{s} = t_{s+1} - t_{s}$ likewise $\psi$. Note that in case of stepwise stress histories the integral is evaluated exactly.

This forward step can be compared with forward integration, see [2.64]. A backward integration step is taken when the stress and Schapery parameters equal the values at the end of the step: $G_{s}^{2}(\sigma) = G_s^{2^{s+1}}$ and $a_{\sigma}(\sigma) = a_{\sigma}^{s+1}$. If the average of the forward and the backward step is taken, then integration corresponding with the trapezium rule is performed. In general:

$$q^{s+1}_{k} = q_{k}^{s} + (1 - \alpha) \left( D_{\Delta} G_{s}^{2} - q_{k}^{s} \right) \left( 1 - \exp \left( - \frac{\Delta t_{s}}{\tau_{s} a_{\sigma}^{s+1}} \right) \right) +$$

$$+ \alpha \left( D_{\Delta} G_{s}^{2^{s+1}} - q_{k}^{s} \right) \left( 1 - \exp \left( - \frac{\Delta t_{s}}{\tau_{s} a_{\sigma}^{s+1}} \right) \right) \tag{2.68}$$

with $\alpha$ the stepping parameter. Since the stress and the Schapery parameters at the end of the integration step depend in a nonlinear manner on the step itself, an iteration process is needed to find the solution if $\alpha > 0$. In this case the step is an implicit integration step. Only the forward step with $\alpha = 0$ can be evaluated directly without iterations and is therefore called an explicit integration step. Numerical instabilities during FEM calculations can occur when the time integration is performed with $\alpha < \frac{1}{2}$, [5.65, 5.66].

Relaxation formulation

The Schapery model in relaxation form is written as:

$$\sigma(t) = E_{0} \theta_{0}(e) + h_{l}(e) \sum_{l} r_{l}(t) \tag{2.69}$$

with for the $l$-th Maxwell element the nonlinear convolution integral:

$$r_{l}(t) = E_{l} \int_{0}^{t} \left( - \frac{\rho_{l}(\xi)}{\tau_{l}} \right) dH_{l}(e) d\xi \tag{2.70}$$

The strain reduced times are defined conform $\psi$:

$$\rho(t) = \int_{0}^{t} \frac{d\xi}{a_{l}(e)} \quad \rho(\xi) = \int_{0}^{\xi} \frac{d\xi}{a_{l}(e)} \tag{2.71}$$

Again for convenience:

$$H_{s}(e) = \delta_{k}(e) \tag{2.72}$$
The next incremental formulation is given without derivation, but can be found by application of the same strategy as followed for the creep formulation:

\[ r_i^{*+1} = E_i H_i(\varepsilon) - \exp\left(-\frac{\Delta \psi_i}{\tau_i} \right) r_i^{*} - E_i \int_{t_i}^{t_i^{*+1}} \exp\left(-\frac{\psi(t_{n+1}) - \psi(t_{n})}{\tau_i} \right) \frac{1}{\tau \alpha_e(\varepsilon)} H_i(\varepsilon) d\xi \tag{2.73} \]

A combined forward/backward integration step gives:

\[ r_i^{*+1} = (1 - \alpha)\left(E_i H_i^* - r_i^*\right) \exp\left(-\frac{\Delta t_i}{\tau_i \alpha_e\varepsilon}\right) + \alpha \left(E_i H_i^{*+1} - r_i^*\right) \exp\left(-\frac{\Delta t_i}{\tau_i \alpha_e^{*+1}}\right) \tag{2.74} \]

The same numerical aspects for the creep form are valid for the relaxation form.

### 2.3.2 Schapery model, differential form

#### Creep formulation

The Schapery model, see equations (2.62) and (2.63), is considered again. Differentiation of \( q_k(t) \) and subsequent substitution of \( q_k(t) \) yields the differential equation:

\[ \frac{dq_k(t)}{dt} = \frac{D_k G_2(\sigma) - q_k(t)}{\tau_k \alpha_\varepsilon(\sigma)} \tag{2.75} \]

This form was applied by Czyz et al. [2.67] and later repeated by Poon et al. [2.68]. A combined forward/backward step gives:

\[ q_k^{*+1} = q_k^* + (1 - \alpha) \left. \frac{dq_k}{dt} \right|_{t_n} \Delta t_n + \alpha \left. \frac{dq_k}{dt} \right|_{t_n^{*+1}} \Delta t_n = \]

\[ = q_k^* + (1 - \alpha) \left( D_k G_2^* - q_k^* \right) \frac{\Delta t_n}{\tau_k \alpha_\varepsilon} + \alpha \left( D_k G_2^{*+1} - q_k^* \right) \frac{\Delta t_n}{\tau_k \alpha_\varepsilon^{*+1}} + \Delta t_n \tag{2.76} \]

Note that a backward step gives \( q_k^{*+1} = D_k G_2^{*+1} \) when \( \Delta t_n \) reaches infinity, whereas the forward step does not. The latter gives overshoot of the equilibrium value \( D_k G_2^* \). Furthermore the differential approach does not result in the exact solution in case of stepwise stress histories.

#### Relaxation formulation

Equations (2.69) and (2.70) are considered. The differential form for the relaxation formulation is less straightforward than for the creep formulation since a strain step is
accompanied by a sudden stress in a Maxwell element whereas a stress step does not result in a strain step in a Kelvin-Voigt element. A linear transformation, applied also by Popelar et al. [2.9], is therefore needed:

$$\tau_i(t) = E_i H_i(t) - \bar{\tau}_i(t)$$  \hspace{1cm} (2.77)

Now the differential form can be derived using the same strategy as in the creep formulation case. A combined integration step gives:

$$\bar{\tau}_i^{n+1} = \bar{\tau}_i^n + (1 - \alpha) \left( E_i H_i^n - \bar{\tau}_i^n \right) \frac{\Delta s_i}{\tau_i a_i^n} + \alpha \left( E_i H_i^{n+1} - \bar{\tau}_i^n \right) \frac{\Delta s_i}{a_i^{n+1} + \Delta t_i}$$  \hspace{1cm} (2.78)

As can be observed the results after the transformation are identical to the creep form in equation (2.76).

2.4 Conclusions

Since creep experiments are very convenient to characterize viscoelasticity the application of a creep model is more straightforward than a relaxation model.

The Generalized Kelvin-Voigt model is capable of describing creep curves which have a complex shape.

For engineering purposes the Schapery creep model seems most adequate to describe the nonlinearity in the time dependent behaviour. This model has been used before for HDPE and it includes the engineering models of Leaderman and Leonov.

The yield behaviour of most thermoplastics can be described by either a single Eyring process or by two Eyring processes in parallel.

The effect of temperature and ageing on creep are commonly described by the principles of time-temperature and time-ageing time superposition, respectively.

The Henriksen method for the numerical evaluation of the response of the Schapery creep model to arbitrary stress histories results in a computationally effective incremental scheme. The method can only be applied if the Generalized Kelvin-Voigt model (a spring dashpot model) is used to describe the creep compliance function. Application of the Generalized Maxwell model in the Schapery relaxation model yields an effective recurrent scheme.
2.5 References


[2.34] Roetling, J.A. *Yield Stress Behaviour of Poly(ethylmethacrylate) in the Glass Transition Region*, Polymer 6, 615, (1965)
Time dependent mechanical behaviour


3 Experimental

3.1 Introduction

The viscoelastic behaviour varies not only with the type of polymer, i.e., chemical structure. Different grades, i.e., molecular structure, of each material are available, each having its own viscoelastic characteristics. Furthermore processing conditions influence the mechanical performance of a polymer through the physical structure. Due to processing the material can become anisotropic, contain internal stresses and, for semi-crystalline polymers, possess a varying degree of crystallinity.

After solidification and cooling down the viscoelastic behaviour can be determined. In general it is still impossible to give an accurate prediction of, for instance, the creep behaviour of a certain polymer when only its chemical and physical conditions are known since only few polymers have been subjected to elaborate testing.

The viscoelastic behaviour of a polymer is often characterized by creep experiments, since these experiments can be performed accurately with long time spans by means of a dead weight. Other characteristics are stress relaxation, recovery after creep and stress response to straining at a constant rate. In case of nonlinear viscoelasticity one single experiment is not sufficient as for linear behaviour. A full characterization of the creep behaviour, for example, then requires creep curves at different stress levels.

Besides a full picture of the stress and strain relations mapping of these relations as a function of other parameters is essential. Particularly the effect of temperature and the effect of ageing are considered important for engineering purposes.

Since it is impracticable to investigate all parameters which influence the viscoelastic behaviour of HDPE only the most relevant have been considered. The influence of the physical structure of HDPE has not been examined, neither was the influence of the processing conditions. However it was decided to perform creep experiments on three different grades of HDPE to get an impression of the extent the creep characteristics change due to different molecular structures.

The other effects that have been considered are the effect of arbitrary stress histories, temperature, ageing and non-tensile stress states, see figure 1.2. Combined effects are not investigated. Creep measurements are considered the reference measurements, since these
are performed most commonly. The phenomenology of the creep data determines which existing models are chosen for the modelling of the nonlinear viscoelastic behaviour.

3.2 Material and specimens

Three different HDPE grades were selected for creep experiments: HDPE 7058Z, HDPE 7108, both supplied by DSM, and HDPE HM5420XP, supplied by Van Leer/BP Chemicals. In table 3.1 some characteristic properties are given. The first two materials are injection moulding grades and the third material is an extrusion HDPE. The properties given are: density \( \rho \), melt flow index MFI, weight average molecular weight \( M_w \), number average molecular weight \( M_n \), modulus of elasticity \( E \) and crystalline volume fraction \( f_c \). For more information on structure related properties see [3.1].

<table>
<thead>
<tr>
<th></th>
<th>7058Z</th>
<th>7108</th>
<th>5420XP</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \rho ) (kg/m(^3))</td>
<td>± 953</td>
<td>± 953</td>
<td>± 955</td>
</tr>
<tr>
<td>MFI (dg/min)</td>
<td>4.4</td>
<td>10</td>
<td>2.1</td>
</tr>
<tr>
<td>( M_w ) (kg/mole)</td>
<td>86</td>
<td>61</td>
<td>355</td>
</tr>
<tr>
<td>( M_n ) (kg/mole)</td>
<td>19</td>
<td>16</td>
<td>20.4</td>
</tr>
<tr>
<td>( E ) (MPa)</td>
<td>± 1100</td>
<td>± 1000</td>
<td>± 1400</td>
</tr>
<tr>
<td>( f_c )</td>
<td>± 58%</td>
<td>± 58%</td>
<td>± 66%</td>
</tr>
</tbody>
</table>

Table 3.1 Material properties of the three different HDPEs.

The HDPE grade 7058Z was taken as the reference material since this grade is also used for injection moulded crates and containers. With this material different geometries were injection moulded: tensile bars, square plates, hollow cylinders and dishes. The injection moulding parameters and product thicknesses are given in table 3.2. Furthermore all the different injection moulding products were processed with similar parameters. After production the specimens were stored at 23°C.

Variations in the degree of crystallinity of HDPE through the thickness of the products are only present in a thin skin layer of approximately 200 \( \mu \)m [3.2, 3.3]. Therefore the main contribution to the mechanical behaviour is from the nearly isotropic core layer. The melt and mould temperature were taken relatively high and the cycle times were taken relatively long (slow cooling after injection moulding) to minimize internal stresses and
3.2 Material and specimens

anisotropy. Creep of bars cut parallel and perpendicular to the flow direction from square plates, exhibit relative differences of less than 7%, see section 5.4.2. This indicates that in our case anisotropy is indeed small. In practice, however, injection moulding cycles are much smaller and then larger variations in viscoelastic properties can be found [3.4].

<table>
<thead>
<tr>
<th>Bar</th>
<th>Plate</th>
<th>Cylinder</th>
<th>Dish</th>
<th>Bar</th>
</tr>
</thead>
<tbody>
<tr>
<td>7058Z</td>
<td>7058Z</td>
<td>7058Z</td>
<td>7058Z</td>
<td>7108</td>
</tr>
<tr>
<td>( t_{\text{cycle}} ) (s)</td>
<td>55</td>
<td>60.8</td>
<td>56</td>
<td>52</td>
</tr>
<tr>
<td>( t_{\text{cooling}} ) (s)</td>
<td>35</td>
<td>15.0</td>
<td>20</td>
<td>15</td>
</tr>
<tr>
<td>( t_{\text{injection}} ) (s)</td>
<td>2.6</td>
<td>12.5</td>
<td>14</td>
<td>6.0</td>
</tr>
<tr>
<td>( p_{\text{injection}} ) (bar)</td>
<td>54</td>
<td>35</td>
<td>28</td>
<td>550±10</td>
</tr>
<tr>
<td>( T_{\text{nozzle}} ) (°C)</td>
<td>250</td>
<td>215 (melt)</td>
<td>250</td>
<td>250</td>
</tr>
<tr>
<td>( T_{\text{cool. water}} ) (°C)</td>
<td>40</td>
<td>62-63</td>
<td>40</td>
<td>40</td>
</tr>
<tr>
<td>( h_{\text{average}} ) (mm)</td>
<td>3.8</td>
<td>3.9</td>
<td>2.7</td>
<td>3</td>
</tr>
</tbody>
</table>

Table 3.2 Injection moulding parameters of the different test products.

![Diagram](image)

Figure 3.1 Tensile bar according to ISO 527; dimensions in mm; dashed areas are clamped.

Bar
The 3.8 mm thick flat tensile bars, see figure 3.1, are according to ISO 527, except that the radius is slightly smaller. The dog bone shape results in a 50 mm section in the middle where the stress state is uniaxial and homogenous. These bars are designed such that 1 mm corresponds to 1% strain in case of linear elasticity and an initial grip to grip separation of 115 mm. The bars were used for creep, creep and recovery, tensile and compressive
straining, stress relaxation and ramp loading experiments in case of HDPE 7058. The 7108 bars were only used for creep, see table 3.4 for the total experimental setup.

**Plate**
The 3.9 mm thick flat square plates with a length and width of 117 mm were injection moulded with a film gate at DSM. The film gate results in less anisotropy and internal stresses compared with a single point gate. The plates were used for tensile creep experiments on smaller bars cut from the plates, see figure 3.2, and for three-point bending creep on prismatic beams which were machined from the middle of the plates and later microtomed to a width of 3.9 mm, see figure 3.3. As a result the beams have a square cross section with the height and width equal to the thickness of the square plates. Bars and beams were cut from the plates both parallel and perpendicular to the injection moulding direction.

![Diagram of plate and bar dimensions](image)

**Figure 3.2** Small tensile bar machined from square plates; dimensions in mm; dashed areas are clamped.

![Diagram of prismatic beam dimensions](image)

**Figure 3.3** Prismatic beam machined and microtomed from square plates; dimensions in mm.

**Cylinder**
Hollow cylinders with a length of 77 mm were injection moulded. The outer diameter measured 11.2 mm and the thickness 2.6 mm (increasing slightly to 2.7 mm at the ends).
See figure 3.4. The cylinders were subjected to creep in tension and to creep in torsion.

**Figure 3.4** Hollow cylinder; dimensions in mm.

**Dish**
Finally axisymmetric dishes were injection moulded. The thickness varies from 3.0 mm in the center to 3.2 near the ridge and then decreases again to 2.1 at the outside of the dish. The diameter of the dish was 151 mm. See figure 3.5 for the radial cross section. The response of the dish to compression with constant speed was examined.

**Figure 3.5** Radial cross section of the dish; dimensions in mm.

**Extruded pipe**
Tensile bars ISO 527 were machined in the length direction from an extruded pipe with a 3.5 mm wall thickness. For the extrusion conditions see table 3.3. Bars machined from the cylinder were not perfectly flat in length direction because of internal stresses frozen in during the extrusion process. The center deviated approximately 2 mm from the straight line between the specimen ends. Also small material variations in the circumferential direction due to the material flow around the fixations of the kernel can be present and introduce extra scatter in the data. The position of the bars in the extruded tube were not registered. The specimens were tested for their creep response only.
<table>
<thead>
<tr>
<th>Output</th>
<th>Pipe 5420XP</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_{\text{extrusion}}$ (bar)</td>
<td>410</td>
</tr>
<tr>
<td>$T_{\text{melt}}$ (°C)</td>
<td>215</td>
</tr>
<tr>
<td>$T_{1 \text{ meter}}$ (°C)</td>
<td>65</td>
</tr>
<tr>
<td>$T_{3 \text{ meter}}$ (°C)</td>
<td>35</td>
</tr>
<tr>
<td>Diameter (mm)</td>
<td>150</td>
</tr>
<tr>
<td>$h$ (mm)</td>
<td>3.5</td>
</tr>
</tbody>
</table>

Table 3.3  Extrusion parameters of the pipe.

3.3 Measurements

Except for the creep experiments intended to study the effect of temperature, all experiments were performed at a constant temperature of 23°C. The humidity was disregarded since HDPE is extremely hydrophobic resulting in negligible moisture absorption. In case not mentioned experiments were performed on HDPE 7058Z. For all experiments the effect of ageing during the experiment can be neglected since the experimental times were less than 30% of the material age, i.e., time after injection moulding/extrusion [3.5]. Furthermore all experiments at, for instance, 32 weeks of age were performed within 30% of those 32 weeks of time. See table 3.4 for an overview.

Engineering stress and engineering strain were determined on separate systems. After the raw data were processed, see section 3.4, the stress and strain results were matched on time bases. For all tests the specimens were held at least 10 minutes without load in the grips of the testing equipment before being loaded, to ensure that the specimens had come to "rest". The strain output measured just before the onset of the stress was set as zero level reference. All experiments were performed on "virgin state" specimens and products. In case tests were performed more than once, average results were taken.
<table>
<thead>
<tr>
<th>Reference measurement</th>
<th>Specimen</th>
<th>Loading</th>
<th>Temp. (°C)</th>
<th>Age (weeks)</th>
<th>Grade</th>
<th>Test</th>
</tr>
</thead>
<tbody>
<tr>
<td>bar</td>
<td>constant force</td>
<td>23</td>
<td>32</td>
<td>7058</td>
<td>tensile</td>
<td></td>
</tr>
<tr>
<td>Arbitrary stress history</td>
<td>bar</td>
<td>constant speed</td>
<td>32, (64)</td>
<td>7058</td>
<td>tensile</td>
<td></td>
</tr>
<tr>
<td></td>
<td>ramp loading</td>
<td>23</td>
<td>32</td>
<td>7058</td>
<td>tensile</td>
<td></td>
</tr>
<tr>
<td></td>
<td>const. - zero force</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>constant displ.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>constant speed</td>
<td>43</td>
<td>128</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Temperature</td>
<td>bar</td>
<td>constant force</td>
<td>18, 23, 28, 33, 38, 43</td>
<td>64</td>
<td>7058</td>
<td>tensile</td>
</tr>
<tr>
<td>Ageing</td>
<td>bar</td>
<td>constant force</td>
<td>23</td>
<td>2, 4, 8, 16, 32, 64, 128</td>
<td>7058</td>
<td>tensile</td>
</tr>
<tr>
<td>HDPE grade</td>
<td>bar</td>
<td>constant force</td>
<td>23</td>
<td>32</td>
<td>7058, 7108, 5420XP</td>
<td>tensile</td>
</tr>
<tr>
<td></td>
<td>beam from bar</td>
<td>sinus force</td>
<td>-150...110</td>
<td>DMTA (3-pb)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Non-tensile stress</td>
<td>bar, bar piece</td>
<td>constant speed</td>
<td>32</td>
<td>tensile, compr.</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>bar from plate, beam from plate</td>
<td>constant force</td>
<td>32, (48)</td>
<td>Tensile, 3-point bending</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>cylinder</td>
<td>constant force</td>
<td>32</td>
<td>tensile, torsion</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>dish</td>
<td>constant speed</td>
<td>32</td>
<td>compr./ bending</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 3.4 Schedule of the experiments.
3.3.1 Creep

Creep on HDPE 7058Z at 23°C was taken as the reference measurement, see the row of "reference measurement" in table 3.4. Since products like containers and bottle crates can be loaded for years during service, the creep response must at least be known up to creep times of a few months. Extrapolation of the data for one decade can be done without much loss in accuracy. A total creep time of 8 weeks was chosen and experiments were started 32 weeks after injection moulding. All experiments were performed within 10 weeks.

Long creep times and an economic use of test facilities could only be accomplished by dead weight loading of the specimens. On the other hand reliable data for short creep times require the use of hydraulic testing machines. Turner [3.6] showed that the measured strain response gives an accurate representation of the exact step response after a time which is 10 times the time needed to apply the constant stress.

The creep experiments were divided into three test series:

Creep test series 1
Fast creep experiments were performed on a Zwick hydraulic machine at the Center of Polymers and Composites of the Eindhoven University. Since the constant force could be applied within 0.03 seconds, measurements are reliable from 0.3 seconds on. The strain was measured with an Instron extensometer with a gauge length of 50 mm. All measurements were carried out twice. The room temperature varied slightly between 22 and 24°C.

Creep test series 2
The second test series was carried out in the Laboratory of Mechanical Reliability at the Delft University. The Zwick electro-mechanical test machine (spindle drive) could apply the creep force within 1 second. The strain was measured with an MTS extensometer with a gauge length of 25 mm. All tests were performed three times at an ambient temperature of 23 ±0.3°C.

Creep test series 3
Dead weight experiments were performed at the Laboratory of Materials Science at the Delft University. The strain was measured with an Instron extensometer with a gauge length of 50 mm (1-6 MPa), 25 mm (8 MPa) and 12.5 mm (10-12 MPa). Tests at 2, 4 and 8 MPa were done twice; the other tests once. Strain measurements were considered accurate from 10 seconds onwards. The temperature was controlled 23 ±0.5°C.
3.3.2 Arbitrary stress history

Different types of experiments were performed with a stress varying in time, see the row of “arbitrary stress history” in table 3.4. The electro-mechanical Zwick was used for these tests and the set up was identical to that of the second test series of creep experiments in section 3.3.1. The test were carried out in duplicate. The only exception are three series of creep recovery experiments which were performed with dead weight loading, which were removed after a certain creep time. Here the test conditions were similar to those of the third creep test series of section 3.3.1. In case not mentioned tests were carried out 32 weeks after injection moulding at a temperature of 23°C.

Tensile straining
During these experiments the grip to grip distance was increased with a constant speed. As a result the strain rate obtained from extensometer measurements will be almost constant. Displacement speeds were taken 1, 0.1 . . . . 0.0001 mm/s resulting in strain rates of approximately 0.01 . . . . 0.000001 1/s. Because of lack of time the slowest straining test was performed 64 weeks after injection moulding.

An extra set of straining experiments were carried out at 43°C and 128 weeks after injection moulding. Strain rates were: 0.1, 0.0316, 0.01, 0.00316 . . . . 0.000000316 1/s. These experiments were carried out once, since they were to be used only for qualitative analysis of the yield behaviour.

Ramp loading
During ramp loading experiments the stress was increased linearly with time. Three stress levels were tested: 4, 8 and 12 MPa. These stress levels were reached in half an hour starting from zero stress. After these stress levels had been reached, the load was removed again with the same speed, resulting in a total experimental time of one hour.

Creep recovery
In these experiments the stepwisely applied constant load was also removed stepwisely after a certain creep time. This stage after creep is called recovery. The recovery time was taken ± five times the creep time. Creep times varied: 150, 300, 600, 1200 and 2400 seconds for electro-mechanical Zwick experiments and 4800, 9600 and 19200 seconds for dead weight creep recovery tests. Load levels were taken 1, 2, 4, 6 . . . . 12 MPa.

Multiple creep recovery experiments were also carried out at stress levels of 4, 8 and 12 MPa. A creep recovery loading history with a creep time of 150 seconds and a five times larger recovery time was immediately followed by four more creep recovery sequences. For each sequence of creep recovery the creep and recovery times were doubled.
Stress relaxation

The moving grip was given a stepwise displacement. As a result the strain measured with
the extensometer was almost constant. These tests were performed 128 weeks after injection
moulding. Displacements of 0.5, 1, 2 . . . . 16 mm were held for 2 hours, leading to strain
levels of approximately 100 times smaller. The experiments with displacements of 0.5 and 1
were also performed with a duration of approximately 20 hours.

Since the electro-mechanical Zwick could apply a stepwise displacement with a
much higher speed (500 mm/min) than a stepwise force, as was the case for creep, reliable
data were obtained at short times already. The time to apply a step displacement equals the
prescribed displacement divided by the speed.

3.3.3 Creep at different temperatures

Tensile bars were subjected to a dead weight loading at the following temperatures: 18, 23,
28, 33, 38 and 43°C, see also the row of “temperature” in table 3.4. The first series of
experiments, performed at 43°C, started 64 weeks after the injection moulding of the bars.
The second series, at 38°C, started 65 weeks after injection moulding, etc. Each experiment
lasted 1 week. Experiments were performed similar to the third creep test series described in
section 3.3.1, except with some variations in the strain gauge length. Exceptions were: 25
mm for 6 MPa at 38 and 43°C and 10 MPa at 18°C, 12.5 mm for 8 MPa at 38 and 43°C, 50
mm for 8 MPa at 18°C.

3.3.4 Creep at different ages

The influence of the material age, i.e., time after injection moulding, on creep was
investigated for the load levels of 2, 4, 8 and 12 MPa, see the row of “ageing” in table 3.4.
Two hour creep experiments were therefore performed on the electro-mechanical Zwick at
ages of 2, 4, 8, 16 . . . . 128 weeks. Experiments were carried out as the second creep test
series of section 3.3.1. Experiments at ages of 2 and 4 weeks were performed twice, all
others three times.

3.3.5 Creep of different grades of HDPEs

The creep experiments on HDPE 7058 were described in section 3.3.1. The second and the
third creep test series as described above, were also performed on injection moulded tensile
bars of HDPE 7108 and on tensile bars cut from extruded pipes of HDPE HM5420XP, see the row of “HDPE grade” in table 3.4. Again all experiments were carried out 32 weeks after injection moulding/extrusion. The bars machined from the pipes were not exactly straight, see extruded pipe in section 3.2. The extensometer was always mounted on the inner side of the curved bars.

Small prismatic beams were machined from the center of the tensile bars of all three grades. The beams measured 2 mm in height and 3.5/3.6 mm in width (=thickness of the center of the bars). The beams were subjected to three-point bending DMTA tests at the Laboratory of Polymer Materials and Engineering. The length between the outer supports was 20 mm and the 1 Hz sinusoidal dynamic load with a minimum to maximum force ratio of 11 was controlled to produce a displacement amplitude of 25 µm. The tests were performed at a temperature increasing from -150°C up to the melt temperature of ±110°C with steps of 1°C. The results of these experiments were used qualitatively only.

3.3.6 Non-tensile stress states

Four types of experiments with non-tensile stress states were performed: compressive straining of cubes, three-point bending of prismatic beams, torsion of hollow cylinders and compression of dishes, see also the row of “non-tensile stress states” in table 3.4. The second and third types of experiments were creep experiments, while in case of the cubes and dishes a constant compressive displacement rate was applied, since creep was hard to accomplish. FEM predictions of the responses, during these experiments with a material model based on tensile creep results of the injection moulded bars, might deviate due to material differences between the bars, plates, cylinders and dishes. Therefore bars from the plates and cylinders were first subjected to tensile creep experiments. In case of the dish this could not be accomplished because of its curved shape.

Compressive straining
Small pieces with 1 cm length were machined from the middle of the injection moulded tensile bars. These cubes were compressed in length direction between two flat smooth plates with constant speeds of 0.1, 0.01 . . . 0.0001 mm/s. Experiments were performed in duplicate at 23°C, 32 weeks after injection moulding. Both specimen ends were greased with a film of vaseline to decrease the effect of lateral friction due to Poisson’s effect. No extensometer could be used, and therefore the displacement of the moving plate was used to obtain strain values. Since perfect contact between the upper specimen end and the upper smooth plate is not reached instantly but gradually, the determination of the strain is somewhat inaccurate for small strain values.
Three-point bending of a prismatic beam
Small tensile bars were cut from 117 mm square plates, see figure 3.2. These bars were cut out of the center of the plates parallel to the injection moulding direction and perpendicular to it. Tensile creep tests of 2 hours were carried out in duplicate on these specimens 32 weeks after injection moulding of the plates at stress levels of 2 MPa . . . . 18 MPa with steps of 2 MPa. Conditions were identical to the second test series of creep experiments in section 3.3.1 except for the different grip to grip distance of 82 mm.

Prismatic strips were machined from the middle of the plates, again both parallel and perpendicular to the injection moulding direction. Later these strips were microtomed to a width of 3.9 mm, see figure 3.4. Consequently prismatic beams with a square cross section were obtained, since the height of the strips was also 3.9 mm, i.e., the thickness of the square plates. These beams were loaded with a dead weight of 600 gram for 2 hours while the displacement of the center was measured. In figure 3.6 the configuration is shown. Bending was performed both out-of-plane of the plate and in-plane. Tests were carried out in duplicate 48 weeks after injection moulding at a temperature of 23 ±0.5°C.

![Diagram](image)

**Figure 3.6**  Three-point bending according to ISO 889-2: dimensions in mm.

Torsion of a hollow cylinder
Before the hollow cylinders, see figure 3.4, were tested in torsion creep, 2 hour tensile creep experiments on the electro-mechanical Zwick were performed in duplicate at 23 ±0.3°C and 32 weeks after injection moulding. Stress levels were: 1, 2, 4, 6 . . . . 16 MPa. Specimens were clamped between a flat clamp and a clamp with a wedge. The hollow center of the cylinders was filled with two metal cylinders at the ends to prevent them to crush during clamping. Since these metal cylinders were fixed to the clamps as well, the creep load could also be transferred to the specimen via the inner surface. The grip to grip distance was 60 mm and the strain was measured in the middle with an MTS extensometer with 25 mm gauge length. The stress and strain were assumed to be homogeneous in the section of measurement.
After the tensile creep experiments, the torsion creep experiments were carried out. Again the hollow centers at the ends were filled, but now the clamping was done with a circular crown which was pressed on the cylinder by a nut. The grip to grip distance was 50 mm. The angle of twist of the moving clamp was monitored during 1 hour of creep. The average torques of the duplicate experiments were: 0.387, 0.787, 1.18 and 1.53 Nm. Temperature control was less accurate: 23 ± 1°C.

Compression of a dish
The dish described in section 3.2, see also figure 3.5, was compressed at the top in the center by a 12 mm diameter cylinder. The rounding off radius of the cylinder end that pressed the dish surface was 40 mm. The brim of the dish was put on a flat metal base and was also surrounded by a metal ridge, suppressing the displacements of this edge. Experiments were performed 32 weeks after injection moulding with compression speeds of 1, 0.1, 0.01 and 0.001 mm/s while the force on the dish was monitored. Tests were carried out twice and the temperature was controlled at 23 ± 0.3°C.

3.4 Data processing
The strain during creep is monitored in time. Since the strain rate is decreasing in time, more measurements are needed at the beginning of the creep curve. Figure 2.9 shows a creep curve with equal spaces between the measurements on logarithmic time scale. Since stress and strain were recorded on separate systems, no data at the specific time points shown in figure 2.9 are available.

To overcome this problem a small interval of the measured creep curve was approximated by a linear regression line. The value of this regression line at the specific point in time can be regarded as a good approximation of the actual strain value. Furthermore the slope of the regression line corresponds to the strain rate. A side effect is that noise on the data is reduced by this method.

A longer time interval will result in a better reduction of the noise in the data. On the other hand too long time intervals invalidate the regression line approximation. The optimal time interval was estimated as follows. Assume power law creep behaviour:

\[ \varepsilon(t) = at^n \]  

(3.1)

In case of creep on HDPE the value of \( n \) varies between 0.07 and 0.25. Elastic strains do not alter the next argument. If now the strain on the interval \( t(1-\Delta) \) to \( t(1+\Delta) \) with \( \Delta = \Delta t/t \)
is approximated by a straight line through the limits of the time interval then the strain becomes:

$$
\varepsilon(t) = \frac{at^n(1-\Delta)^n + at^n(1+\Delta)^n}{2} = at^n\left(1 + \frac{1}{n} n(n-1)\Delta^2 + O(\Delta^4)\right)
$$

(3.2)

The strain rate becomes:

$$
\frac{d\varepsilon(t)}{dt} = \frac{at^n(1+\Delta)^n - at^n(1-\Delta)^n}{2t\Delta} = ant^{n-1}\left(1 + \frac{1}{n} (n-1)(n-2)\Delta^2 + O(\Delta^4)\right)
$$

(3.3)

If the interval parameter $\Delta = 0.15$ then the maximum relative errors become -0.002 and 0.007 for the strain and strain rate, respectively. Since the regression line also takes into account data closer to time $t$, errors will even be smaller. During the secondary creep stage no errors will be present. Therefore strains during creep were obtained by regression analysis of the raw data with time interval parameter $\Delta = 0.15$.

The same procedure was applied for recovery, ramp loading and stress relaxation experiments. In case of constant strain rate experiments the strain data were determined on linear time scale. The time interval for the regression line approximation was taken from $t - \Delta t$ to $t + \Delta t$ with $\Delta t$ the time step between the data points. The strain and strain rate are then determined very precisely since the strain time function is (almost) linear.

### 3.5 Accuracy

The measurements can be regarded as representative when systematic errors and scatter in the data are less than a few percent. Systematic errors are the result of incorrect assumptions, and since the errors are repeated consistently during the measurements, they cannot be found easily by consideration of the data because they do not cause scatter. Scatter in the data is the result of inconsistent, i.e., varying procedures during experimenting.

**Systematic errors**

All extensometers and load cells were checked before use during experiments. Furthermore special care was addressed to the alignment of specimens and clamps. Slip in the clamps during the torsion experiments will invalidate the experiments since no extensometer was used. However slip was not found. Friction during dead weight three-point bending experiments was minimal. The strain data obtained by compression are somewhat unreliable because of the gradual contact between specimen and moving plate. The result of this is a strain shift of ±0.006, see figures 5.1 and 5.2. Also lateral friction preventing the thickening
caused by the Poisson effect could be present.

The creep measurements on the bars from the extruded pipes contain a small systematic error because the bars were slightly curved, see extruded pipe in section 3.2. There are systematic differences between the two test series, since the specimens were clamped in the electro-mechanical Zwick but not during dead weight creep experiments where hooks were put in holes at the ends. Relative deviations are up to 20%, see figure 4.66. This results in different curves during zero load. The results of clamped specimens can be considered more accurate, since part of the curvature is oppressed by the clamping.

Scatter
The most important causes of scatter in the measurements are:
1) Scatter in the material properties due to the production process. Injection moulded specimens were only used from the moment that the injection moulding process had stabilized. Extrusion of a pipe will result in material variations with the circumferential direction due to the flow around the fixations of the kernel. Creep measurements on the extruded pipe can therefore contain more scatter.
2) Inaccurate temperature control. As can be seen in section 4.4 the influence of the temperature on the behaviour of HDPE is large. Variation in the temperature during testing will lead to scatter. Temperature was measured before and after each experiment.
3) Human errors: scatter in positioning of the extensometer. Positioning of the extensometers was done with great care though variations were inevitable. The result is that the knives of the extensometer could twist or slip slightly in the beginning of load application. Therefore small "elastic-like" strain errors could occur. This effect will have a relatively smaller effect at higher strain levels.

3.6 Conclusions

A wide variety of experiments were carried out. The effects of a varying stress history, temperature, ageing, HDPE grade, and non-tensile stresses were included.

In practice the injection moulding cycle times are much smaller than in our case and the effect of injection moulding conditions on the viscoelastic properties can be relatively large.

Engineering stress and strain data were processed in such a way that the level of noise is decreased and that strain rate data are obtained as well.
The creep results on bars cut from extruded pipes are less accurate. Also the strain values measured during compression of cubes are less accurate at small strain levels. Scatter in the measurements was especially caused by temperature variations and small "elastic strain" errors due to the extensometer fixation.

3.7 References


4 Constitutive modelling

4.1 Introduction

Three nonlinear viscoelastic models were evaluated for their capability to fit our experimental creep data on HDPE 7058. In case not mentioned, all data were obtained at 23°C. Through application of models with an increasing degree of complexity, improvements of the description of the creep data were accomplished. The creep curves were fitted with the Schapery creep model, the two-processes model and the Schapery relaxation model. Where the Schapery creep model is an engineering model the two-processes model is more complex since it assumes that two nonlinear viscoelastic processes determine the viscoelastic behaviour. The Schapery relaxation model assumes that the nonlinearity depends on the strain instead of the stress as the Schapery creep model does. This makes fitting of the creep curves more difficult.

With the Schapery creep model the creep curves could be fitted up to 5% strain. The other two models could describe creep of HDPE up to the maximum strain level applied; i.e., 20%. From predictions of the models for stress histories other than constant creep stress it was concluded that the response to stress histories with a monotonous stress variation in time could be predicted properly, and that predictions for non-monotonous stress histories were unreliable due to the occurrence of viscoplasticity. The Schapery creep model was extended to include viscoplasticity.

Although HDPE turned out to be thermo-rheologically complex, up to strain levels below the yield points, i.e., where the Schapery creep model is valid, the effect of temperature could be described by horizontal and vertical shifting of a master curve. The effect of ageing was described by time-ageing time superposition. However vertical shifts were again needed. Also the creep curves of three different grades of HDPE were compared. Stress states other than the uniaxial tensile stress state are discussed in chapter 5.

4.2 Creep

Figure 4.1 shows the engineering strain during our creep experiments at 23°C as markers. The experimental set up is given in section 3.3.1. The creep strain was measured with an
extensometer during three test series, denoted here with numbers 1, 2 and 3.

The creep curves corresponding to stresses below 10 MPa exhibit a pronounced convex curvature at approximately 4000 seconds. Such a convex curvature was also found by Turner for PP [4.1] and for HDPE [4.2]. Also the creep curves of Cessna [4.3] for PP show qualitatively the same behaviour as our HDPE. Struik [4.4] found a convex curvature in the creep curves of LDPE. The stress relaxation curves of HDPE measured by Popelar et al. [4.5] show a pronounced curvature (concave) at times between 1000 and 10000 seconds, independent of the applied strain level.

![Image](image.png)

**Figure 4.1** Strain during creep experiments at load levels of 1 (v), 2 (●), 4 (○), 6 (●), . . . 18 (○) MPa at 23°C.

Figure 4.1 shows that the creep curves belonging to stresses above 8 MPa deviate from the pattern found at lower stress levels. The upward curvature found at stresses above 10 MPa corresponds to a fast decreasing stiffness. The large slope (1 decade right, 1 decade up) of the creep curves at strain levels of 20% implies that the strain rate is almost constant (secondary creep stage) and that the material starts to yield. This can also be derived from the strain rate curves of figure 4.5. The yielding will eventually lead to failure of the
material. During the 10 MPa dead weight creep experiment, cracking occurred at the knife edge of the extensometer before yielding.

![Graph showing creep isochrones](image)

**Figure 4.2** Creep isochrones obtained from the creep data at creep times of 1 (◇), 10 (○), 100 (△) . . . . 1000000 (▽) s.

The stress-strain relation of the creep isochrones in figure 4.2 are obtained by consideration of the creep curves at given creep times. The curvature of the creep isochrones indicates that the creep behaviour of HDPE is nonlinear. Also at longer creep times the nonlinearity is more pronounced. This means that at short creep times a linear approximation is accurate up to higher stress levels than at long creep times. The creep modulus, which is the creep stress divided by the creep strain, is shown in figure 4.3. The creep modulus is the reciprocal of the creep compliance, which is given at the second y-axis. The creep modulus curves are situated closer together at short creep times, also indicating that the creep behaviour is more linear at short times.

Since the pronounced convex curvature occurs at a creep time of 4000 seconds independent of the stress level, time-stress superposition, i.e., horizontal shifting of a master
curve on logarithmic time scale, see section 2.2.2, seems inappropriate to describe the nonlinearity in the creep behaviour. Vertical shifting looks more promising.

![Graph showing creep modulus and compliance curves](image)

**Figure 4.3** Creep modulus (creep compliance) curves obtained from the creep data at load levels of 1 (♦), 2 (♦), 4 (♦), 6 (♦), \ldots 18 (♦) MPa.

### 4.2.1 Schapery creep model

The 1 to 8 MPa creep curves of figure 4.4 do not superimpose completely by a vertical shift since the curves are situated closer together at short creep times than at long creep times. However the creep strain rate curves in figure 4.5 can be superimposed upon a master curve by vertical shifting. The viscoelastic creep strain can therefore be evaluated by multiplication of a time function and a nonlinear stress function, i.e., separation of variables.

The HDPE creep data of Lai et al. [4.6] were fitted by horizontal and vertical shifting of a master curve. Horizontal shifts were up to 5 decades. However the maximum creep time of their data was 1000 seconds, which is situated before the convex curvature. Furthermore when Lai et al. [4.7] described creep and recovery data of HDPE, the horizontal shifts were at maximum $\frac{1}{3}$ decade only.
4.2 Creep

![Graph of creep strain vs. creep time](image)

**Figure 4.4** Strain during creep experiments at load levels of 1 (○), 2 (●), 4 (○), 6 (●), 10 (○), and the true stress/strain Schapery creep model fit (solid lines).

By disregarding horizontal shifting \(a = 1\) and the pre-integral function \(g_0 = 1\) and by assuming that the instantaneous strain is a linear function of the stress \(g_0 = 1\), the following expression for the creep strain is obtained, see equation (2.29):

\[
\varepsilon_i(t) = D_0 \sigma + \Delta D(t) \sigma g_5(\sigma)
\]  

(4.1)

The model for the transient creep strain therefore reduces to the Leaderman creep model, see equation (2.22). The transient creep compliance function \(\Delta D(t)\) was approximated with the Generalized Kelvin-Voigt model, see section 2.2.1:

\[
\Delta D(t) = \sum_k D_k \left(1 - \exp\left(-\frac{t}{\tau_k}\right)\right)
\]

(4.2)

with 11 elements with \(\tau_k = 10^k; \ k \in \{-2, -1, \ldots, 8\}\). The Leaderman (Schapery integral) function \(g_5(\sigma)\) was set equal to a fourth order polynomial with \(g_5(0) = 1\).
Figure 4.5 Strain rate during creep experiments at load levels of 1 (▼), 2 (☆), 4 (○), 6 (●), …, 18 (◇) MPa and the true stress/strain Schapery creep model fit (solid lines).

Since the true stress/strain can only be approximated by the engineering stress/strain up to strain levels of 0.02, creep strain data above this strain level were not considered during the first fit. $D_0$, $D_k$ and $c_i$ in $g_2 = 1 + c_1 \sigma + c_2 \sigma^2 + c_3 \sigma^3 + c_4 \sigma^4$ were used to minimize the relative quadratic errors of the strain and the strain rate, i.e., fitting. The minimization was done with Microsoft Excel using Solver with the options: quadratic estimates, central derivatives, and conjugate search.

The Schapery creep model could be fitted accurately to the creep data. The creep strains of the model are partly shown in figure 4.6 as solid lines. Figure 4.7 shows the values of $g_2(\sigma)$ as diamonds for stresses of 1, 2, 4, …, 12 MPa; i.e., the creep stress levels to which was fitted. An attempt to describe the creep results up to strain levels above 0.02 was made by basing the Schapery model on true stress/strain.
4.2 Creep

Figure 4.6  Strain during creep experiments at load levels of 4 (○), 6 (●), … 18 (♦) MPa and the engineering stress/strain Schapery fit based on engineering stress/strain (solid lines) and based on true stress/strain (dashed lines).

Figure 4.7  Leaderman (Schapery integral) function $g_2(\sigma)$ of the engineering stress/strain fit (♦) and the true stress/strain Schapery creep model fit (solid line).

Implementation of true stress/strain
The maximum strains during the creep measurements were just below 20%. These strain levels no longer allow engineering stresses and strains to be used. Instead true stresses and true strains had to be implemented. The true cross-sectional area, required for the true stress, was determined with the assumption of constant volume. Lai [4.8] found Poisson’s ratios of approximately 0.45 for HDPE. At 10% strain the relative difference between the true stress for a Poisson’s ratio of 0.45 and the true stress for a ratio of 0.5 is only 1%. The assumption
of constant volume therefore will lead to small deviations only.

Equation (4.1) now gives the response to a constant true stress in terms of true strain. Because of the increasing strain during creep the cross-sectional area is decreasing and consequently the true stress is increasing in time. The response to a constant nominal stress (constant force) then needs to be calculated numerically. To do so a time decade was split into 32 time steps which were equidistantial on logarithmic scale. The strain was determined by backward integration of the differential form, see equation (2.76):

\[ q_{k}^{n+1} = q_{k}^{n} + (D_{k} g_{k}^{n+1} \sigma_{n+1} - q_{k}^{n}) \frac{\Delta t_{n}}{\tau_{k} + \Delta t_{n}} \]  

(4.3)

with the total strain equal to:

\[ \varepsilon_{n+1} = D_{0} \sigma_{n+1} + \sum_{k} q_{k}^{n+1} \]  

(4.4)

The true stress was determined in a forward manner with respect to the cross-sectional area; the true strain determination is direct:

\[ \sigma_{true}^{n+1} = \sigma_{eng}^{n+1} (1 + \varepsilon_{eng}^{n}) \] \[ \varepsilon_{true}^{n+1} = \ln(1 + \varepsilon_{eng}^{n+1}) \]  

(4.5)

This method did not suffer from any numerical instabilities. Furthermore by increasing the number of time steps to 64 per decade, the calculated strain changed with less than a percent only.

---

**Figure 4.8**  Retardation spectrum (Kelvin-Voigt parameters) found by the true stress/strain Schapery creep model fit. \((D_{0} = 0.000629 \text{ I/MPa})\).
The fitting parameters of the engineering stress/strain Schapery creep model fit were also used in the model in terms of true stress and true strain to evaluate the creep response. The results are given together with the engineering results in figure 4.6. The figure shows that deviations from the engineering stress/strain approach of the beginning of this section occur from 0.02 strain and are no longer negligible for strain levels above 0.04.

The creep data below 5% strain were considered for the new fit with the Schapery creep model based on true stress/strain. The same fitting procedure as with the engineering fit was applied. The results are shown in figures 4.4 and 4.5. The Leaderman function \( g_2(\sigma) \) and the retardation spectrum are given in figures 4.7 and 4.8, respectively. Numerical values are given in appendix A. The Leaderman function is now a continuous function, since the creep stress levels are no longer fixed but are a function of the creep time.

The fit is consistent with the creep data up to strains of 0.05. Above this strain level the strain rate starts to deviate from the pattern found at low strain levels. The strain rates decrease less, stabilize and eventually increase again, leading to an upward curvature. This yielding of the material could not be described by this model. Another model which can describe the yielding is presented in the next section.

### 4.2.2 Two-processes model

To model the yield behaviour two parallel Eyring processes are required for many polymers, see section 2.2.3. This is not only the case for ultra high modulus PE fibers [4.9]. Roetting showed that this also applies for PMMA, PEMA and PP [4.10 - 4.12]. Also for PVC two processes are required [4.13]. This yield behaviour is as follows: At high strain rates two processes contribute to the total stress measured. At low strain rates one process hardly carries any load and the other process alone is responsible for the stiffness. The result is that the yield behaviour at high strain rates or high yield stresses is stiffer.

In figure 4.9 the stress measured during tensile straining at 43°C is shown for a number of strain rates as a function of the imposed strain. When these data are transformed into true stress and true strain the solid lines are obtained. In fact no true yielding occurs since the true stress is increasing constantly. If the true stresses at a true strain of 0.15 during the tensile straining experiments (crosses) are plotted against the true strain rate on logarithmic scale then the Eyring plot of figure 4.10 is obtained. It can be observed that an increase in strain rate at small strain rates results in a smaller increase of the stress than is the case at high strain rates. Apparently at least two parallel processes are responsible for the stress at strain levels of 0.15.
Figure 4.9  Stress during straining experiments at 43°C with strain rates of approximately 0.1 (○), 0.0316 (●), 0.01 (△) . . . 0.000000316 (▲) 1/s. The markers denote measurements and the lines denote the data transformed into true stress/strain.

The yield behaviour found during creep confirms the presence of at least two processes. The creep curves in figure 4.11 at strain levels of 10%, where yielding occurs, lie closer together before the convex curvature, i.e., 12-18 MPa, than after this curvature; i.e., the 10-12 MPa curves. This can also be observed in figure 4.12 where the yield points are the points with minimum strain rates. Again stiffer yield behaviour is observed at higher stress levels. At stress levels larger than 12 MPa a stress increase of 2 MPa gives less increase in the yield strain rate than at stress levels below 12 MPa.
Figure 4.10  Eyreing plot of the true stress at a true strain level of 0.15 during straining experiments at 43°C (crosses) and the fit with two parallel Eyreing processes (solid line). ($\sigma_0^1 = 1.4$ MPa and $\sigma_0^2 = 0.28$ MPa)

Klompen et al. [4.14] numerically simulated the creep response of a hypothetical material with two Eyreing processes in parallel with each process having a complete creep spectrum, see figure 4.13. Both processes behave according to the Schapery creep model of section 2.2.2 with the Eyreing function for time-stress superposition:

$$a_{\varepsilon}(\sigma) = \frac{\sigma/\sigma_0}{\sinh(\sigma/\sigma_0)}$$  \hspace{1cm} (4.6)

The other Schapery functions drop out (Leonov model): $g_0(\sigma) = g_1(\sigma) = g_2(\sigma) = 1$. The results show striking similarities with our creep data. Their creep curves also exhibit a pronounced convex curvature at an almost constant creep time. Also the creep curves at yielding are situated closer together before this convex curvature (two Eyreing processes
active) than after (one Eyring process active).

This model was therefore used to describe our creep data. To do so two Generalized Kelvin-Voigt models were placed in parallel, see figure 2.7. The true stress is equal to the sum of both true stresses while both processes have equal true strains. The volume was assumed to be constant for true stress determination. The response to a constant force was calculated numerically.

\[ q_{k+1} = q_k + (D_k \sigma_{k+1} - q_k) \frac{\Delta t_k}{\tau_k a_{k+1} + \Delta t_k} \]  \hspace{1cm} (4.7)

**Figure 4.11** Strain during creep experiments at load levels of 1 (✓), 2 (✓), 4 (✓), 6 (●) . . . 18 (●) MPa and the two-processes model fit (solid lines).

The true stress was calculated in the same manner as for the Schapery creep model fit using equations (4.5). Again a time decade was split into 32 time steps equidistantial on logarithmic scale. The backward integration scheme was applied, see equation (2.76):
The total strains of both processes were calculated with equation (4.4). However only the total stress at the end of a time step is known and not its distribution over the two viscoelastic processes. Secant’s iteration method [4.15] was used to find the unique stress distribution which yields equal strains for the two processes.

![Graph](image)

**Figure 4.12** Strain rate during creep experiments at load levels of 1 (v), 2 (●), 4 (○), 6 (●) ... 18 (●) MPa and the two-processes model fit (solid lines).

Process 1 was chosen to be the process which drops out at small strain rates. This process therefore has a single plastic dashpot in the Generalized Kelvin-Voigt. Process 2 did not have this dashpot \( \eta_2 = \infty \) since the true stress in figure 4.9 is ever increasing. The retardation times were taken \( \tau_k = 10^k \) with for process 1 \( k_1 \in \{-2,-1,...,6\} \) and for process 2 \( k_2 \in \{-2,-1,...,18\} \). The springs of both processes, \( D_0^1 \) and \( D_0^2 \), and the viscosity of process 1, \( \eta_1 \), were used to fit the data as well as both time-stress superposition functions \( a_0^1(\sigma) \), \( a_0^2(\sigma) \) and both spectra \( D_1^1 \), \( D_1^2 \). The time-stress superposition functions were set equal to the exponent of a 7th order polynomial and the exponent of a 9th order polynomial and the spectra were described on double logarithmic scale by a 2nd and a 6th order polynomial for
process 1 and 2, respectively. The fitting was done as with the Schapery creep model.

The results of the fit are displayed in figures 4.11 and 4.12 as solid lines. For the dashed lines see section 4.3.2. As can be observed the data are described accurately over the entire region including the yield behaviour. The spectra of both processes are shown in figure 4.13 and the time-stress superposition functions in figure 4.14. For numerical values see appendix A.

The spectra of both processes are smooth but the time-stress superposition functions are not. Fitting with the Eyring function for time-stress superposition, equation (4.6), did not result in a good total fit of the data. For comparison, two Eyring function fits to the actual time-stress superposition functions are added. The Eyring values of these fits deviate from those given in figure 4.10. The irregularity of the time-stress superposition functions can have various causes. In section 4.3.2 the two-processes model will be subjected to further investigation.

![Figure 4.13](image)

**Figure 4.13** Retardation spectra (Kelvin-Voigt parameters) of the two processes. ($D_0^1 = 0.00139$ MPa, $D_0^2 = 0.00121$ MPa and $\eta = 2.17 \times 10^5$ MPa.s)

The true stresses of both processes during creep were evaluated and are given in figures 4.15 and 4.16. They show that at short creep times, before the convex curvature, both processes carry load. When the creep time advances the end of the spectrum of process 1 is reached. The strain of this process is then concentrated in the single dashpot. The result
is that at larger creep times the stiffness and stress of process 1 drop, causing the convex curvature. As was pointed out by Klompen et al. [4.14] the vanishing of the stress in process 1 makes it enter the linear viscoelastic region with no time-stress superposition, i.e., horizontal shifting, present any more. This explains why the convex curvature hardly changes its position in time when the creep stress level is altered.

![Graph showing stress-strain relationship](image)

**Figure 4.14** Time-stress superposition functions of the two processes (solid lines) and as comparison two Eyring function approximations (dashed lines; $\sigma_0^1 = 2.1$ MPa and $\sigma_0^2 = 0.60$ MPa)

A convex curvature might seem hard to reconcile with the elimination of process 1 since it means a lower strain rate at large creep times. This can be explained however by consideration of the creep curves backwards in time. At large creep times the creep curve shows the response of process 2 only since the effect of the interference of process 1 at small creep times is vanished a decade after process 1 has dropped out. [4.2]. The response of process 2 alone at small creep times can be approximated by extrapolation of the end of the creep curve to short times. The extrapolated strain is larger than the actual measured strain because of the extra stiffness given by process 1.

The vertical superimposability of the creep strain rate curves, i.e., the starting point of the Leaderman (Schapery) creep model, can, in this case, be described by the two-
processes model. However, this is not necessarily so. After the convex curvature process 2 carries the complete creep load and therefore exhibits time-stress superposition; horizontal shifting. The vertical superimposability only remains valid for long creep times when the creep curve after the convex curvature is a straight line on double logarithmic scale.

![Graph](image)

**Figure 4.15** *True stress in process 1 during creep at 1, 2, 4, 6 ... 18 MPa (up in graph).*

![Graph](image)

**Figure 4.16** *True stress in process 2 during creep at 1, 2, 4, 6 ... 18 MPa (up in graph).*

### 4.2.3 Schapery relaxation model

Popelar et al. [4.5] found that stress relaxation curves of HDPE could be described by vertical shifting on logarithmic stress scale. Their curves also exhibit a pronounced curvature, be it concave, at a relaxation time independent of the strain level. Measurements on our HDPE in relaxation, see section 4.3.2, are in agreement with the measurements of Popelar et al. Since the Schapery relaxation model is far more simple than the two-processes model an attempt to fit the creep data with this model was made.

In analogy to the assumptions for the Schapery creep model, it was assumed that \( h_0 = h_1 = a_e = 1 \), reducing the time dependent part of the model to the Leaderman relaxation model, see equation (2.21). The stress during stress relaxation then becomes (compare equation (4.1)):

\[
\sigma(t) = E_0 e + \Delta E(t) e h_2(e)
\]

(4.8)
Where the transient relaxation modulus function was described by the Generalized Maxwell model, see section 2.2.1:

\[ \Delta E(t) = \sum_{i} E_i \exp\left(-\frac{t}{\tau_i}\right) \]  

(4.9)

with 11 elements with \( \tau_i = 10^l; \ l \in \{-2,-1,...,8\} \). The Leaderman (Schapery integral) function \( h_z(\varepsilon) \) was set equal to the reciprocal of a ninth order polynomial with \( h_z(0) = 1 \). Again the material behaviour was determined with true stress/strain, see equations (4.5).

![Figure 4.17](image.png)

**Figure 4.17** Strain during creep experiments at load levels of 1 (○), 2 (▲), 4 (○), 6 (●), . . . 18 (●) MPa and the Schapery relaxation model fit (solid lines).

The creep response of the relaxation model was calculated numerically dividing a time decade in 32 equal time steps. The stress was determined with the following equations, see section 2.3.2:
\[ \bar{r}_i^{s+1} = \bar{r}_i^s + \left( E_i h_i^{s+1} \varepsilon_{s+1} - \bar{r}_i^s \right) \frac{\Delta t_i}{\tau_i + \Delta t_i} \]  \hspace{1cm} (4.10)

with the total stress equal to:

\[ \sigma_{s+1} = E_0 \varepsilon_{s+1} + \sum_i \left( E_i h_i^{s+1} \varepsilon_{s+1} - \bar{r}_i^{s+1} \right) \]  \hspace{1cm} (4.11)

Since the nonlinearity \( h_i(\varepsilon) \) of the model is a function of the unknown strain and unlike in the creep model of the known stress, an iteration process is needed for the backward integration steps. Secant's iteration method was used to find the unique strain which yields the imposed creep stress as output, see [4.15].

**Figure 4.18** Strain rate during creep experiments at load levels of 1 (v), 2 (v), 4 (o), 6 (s) . . . 18 (o) MPa and the Schapery relaxation model fit (solid lines).

The fitting was done as with the Schapery creep model. The results are given in figures 4.17 and 4.18. As can be observed the overall fit is better than that of the Leaderman
creep model. The creep model fit is slightly better for the low stress levels. This is not surprising however since every creep curve has a different stress level but not completely different strain levels; there is a strain overlap. The value of \( h_1(c) \) must satisfy more creep curves for some strain ranges, which is more difficult.

What can also be observed in figure 4.18 is that at yielding the model response seems somewhat unstable. Decreasing the time step size by dividing a time decade into 128 steps in stead of 32 however did not give an improvement. Similar waves as in figure 4.18 were also present for the other two models and are connected with the quotient of the characteristic times of two successive Kelvin-Voigt or Maxwell elements, i.e., the space between \( \tau_k \) and \( \tau_{k+1} \) on logarithmic scale. However for this relaxation model the waves worsen during yielding because of the large strain increase during yielding. The true stress during yielding only increases with 10 to 20%.

![Graph showing relaxation spectrum](image)

**Figure 4.19** Relaxation spectrum (Maxwell parameters) found by the Schapery relaxation model fit. (\( E_0 = 11.6 \) MPa).

The relaxation spectrum is given in figure 4.19. For numerical values see appendix A. The linear spring \( E_0 = 11.6 \) MPa has such a low modulus that it only plays a role at high strain levels \( \varepsilon \approx 0.2 \) and can be considered an insignificant fitting parameter. By omitting the linear spring the model reduces to the Leiderman relaxation model, see equation (2.20). This implies that stress relaxation curves should shift vertically on logarithmic stress scale, as was measured by Popelar et al. [4.5].

The pronounced convex curvature of the creep curves around 4000 seconds is not easily noticeable in the relaxation spectrum, whereas this was clearly noticeable in the creep spectrum of figure 4.8. It turns out that the convex curvature in the creep curves is caused by
the relatively high modulus of the Maxwell element with the highest characteristic time. The first two elements of the Generalized Maxwell model also deviate from the general straight line of figure 4.18. These deviations only affect the first decade of the creep curves and were caused by the slight concave curvature at the beginning of the creep curves and by the fitting procedure.

![Figure 4.20](image)

**Figure 4.20** Leaderman (Schapery integral) function $h_z(\epsilon)$ of the Schapery relaxation model fit.

The reciprocal of the Leaderman relaxation function $h_z(\epsilon)$ is shown in figure 4.20; $H_z(\epsilon) = \alpha h_z(\epsilon)$, which gives a more comprehensive picture, is shown in figure 4.21. The yielding of the material is described by the flattening of the “excitation” function $H_z(\epsilon)$ at high strain values, see figure 4.21. Although this model cannot describe Eyring type of yielding behaviour, the complex yielding behaviour is described reasonably by the strain softening caused by the flattening of $H_z(\epsilon)$.

The observation that creep curves (almost) shift vertically on logarithmic strain scale is no longer so obvious as for the creep model. Consider equation (2.19) in which case $H(\epsilon) = H_z(\epsilon)$. Vertical shifting of a creep curve on logarithmic strain scale corresponds to a multiplication of the strain, which then corresponds to an increase of the constant stress. This can only be the case if:

$$H_z(\alpha \epsilon) = f(\epsilon)H_z(\epsilon)$$  \hspace{1cm} (4.12)

$H_z(\epsilon)$ must then be a power function: $H_z(\epsilon) = a \epsilon^p$. A double logarithmic plot of $H_z(\epsilon)$ should then yield a straight line. As can be observed in figure 4.21 for the first strain decade, corresponding to creep strain levels where vertical shifting applies, the excitation function
4.2 Creep

$H_2(\varepsilon)$ has a minor curvature only. The curvature is responsible for the fact that the creep curves are further apart from each other at longer creep times, i.e., higher strain levels, see also figure 4.3.

![Graph](image)

**Figure 4.21** "Excitation" function $H_2(\varepsilon) = \varepsilon h_2(\varepsilon)$ of the Schapery relaxation model fit (solid line) and the function for linear viscoelastic behaviour (dashed line).

The straight line given in figure 4.21 corresponds to linear viscoelastic behaviour; $H_2(\varepsilon) = \varepsilon$. Therefore the measured creep behaviour can be considered linear when strain levels are below 0.1%. This implies that the creep behaviour at short creep times is more linear than at long creep times. When the Schapery creep model was used results were similar: a linear elastic strain and a nonlinear viscoelastic strain also result in more linear behaviour at short times.

4.3 Arbitrary stress histories

In the previous sections creep data were fitted with three different models. In this section the validity of the three models is investigated by comparing measurements and model predictions for situations other than creep. First stress histories with increasing stress were considered since stress reversal and especially unloading until zero stress, i.e., recovery, are notorious for their complex and often unpredictable behaviour. Second the behaviour during unloading was investigated.
4.3.1 Monotonous stress histories

Two types of experiments with increasing stress were performed. In the first type a constant speed to one of the tensile specimen ends was applied, resulting in an almost constant strain rate. The tensile straining experiments were performed with different constant values of crosshead speeds, i.e., strain rates. In the second type the stress was increased in half an hour from zero stress up to the maximum stress level. This is called ramp loading. The stress vs. time diagram shows a straight line. For further details on the experiments see section 3.3.2.

The responses of the three models to tensile straining were determined numerically. The strain measured by the extensometer as a function of time was given as input to the three models to evaluate the stress. For this the time interval corresponding to a crosshead displacement of 1 mm and an engineering strain of approximately 1% was split into 16 equal time steps. Since the strain was taken as input, the true stress was determined backwards with respect to the strain. See equations (4.5) for the forward case. Again for all models backward integration of the differential forms was performed. In case of the Schapery creep model and the two process model, the Secant’s method was used for numerical inversion of the model.

For the response to the ramp loading experiments exactly the same equations and methods were used as for calculating the creep responses, see sections 4.2.1 - 4.2.3. The 225 time integration points were taken equidistantial on logarithmic time scale from 0.1 to 1800 seconds.

Schapery creep model

The predictions of the Schapery creep model for tensile straining and ramp loading are shown together with the experimental data in figures 4.22 and 4.23, respectively. The Schapery creep model gives good predictions up to strain levels of approximately 7%. This is no coincidence since the creep data were described properly up to this strain level only. At higher strain levels the material starts to yield and the model is no longer valid. The experimental data from the ramp loading experiments are described accurately by the model. The strain levels during ramp loading remained below 5% and therefore yielding did not play an important role during these experiments. Indeed The deviations at strain values below 0.001 can be attributed to the inaccuracy of the strain measurement at these small values.
Figure 4.22 Stress during tensile straining experiments with strain rates of approximately 0.01 (o), 0.001 (Δ), . . . 0.000001 (Δ) 1/s and the Schapery creep model predictions (lines).

The Schapery creep model fit is used to compare its response to tensile straining (full viscoelastic analysis) with its response to tensile straining when the stress strain relation given by the creep curves are used (analysis with creep isochrones), see figure 4.2. For the full viscoelastic analysis equations (4.3, 4.4) are used. For calculations with creep isochrones the experimental time and stress are substituted in (4.1) to obtain the strain. Both results are given in figure 4.24. As can be observed the stress differences between the two approaches are 15% at maximum.
Figure 4.23  Strain during ramp loading experiments with 4 (○), 8 (△) and 12 (□) MPa reached in half an hour and the Schapery creep model predictions (lines).

Figure 4.24  Comparison of full viscoelastic analyses (solid lines) and analyses using creep isochrones (dashed lines) with the Schapery creep model for tensile straining at 0.01, 0.001 . . . 0.000001 1/s (down in graph).
Figure 4.25  Stress during tensile straining experiments with strain rates of approximately 0.01 (○), 0.001 (△) ... 0.000001 (○) 1/s and the two-processes model predictions using fit 1 (solid lines) and fit 2 (dashed lines).

Two-processes model
With the two process model the creep data could be fitted over the entire strain region, including the yielding behaviour. In figure 4.25 it can be observed that the predictions (fit 1) for tensile straining, given as solid lines, describe the measurements accurately over the entire region. Also the fastest straining experiment, with stress levels far higher than during creep, is described properly. One has to bear in mind that the creep data contain only few measurements of the yielding process, whereas the tensile straining data contain many. Figure 4.26 shows the two-processes model predictions for ramp loading together with the data. Again predictions are in agreement with the data. The dashed lines will be discussed in section 4.3.2.
Figure 4.26 Strain during ramp loading experiments with 4 (○), 8 (∆) and 12 (□) MPa reached in half an hour and the two-processes model predictions using fit 1 (solid lines) and fit 2 (dashed lines).

Schapery relaxation model
The Schapery relaxation model gives predictions for tensile straining, see figure 4.27, which in total are better than the creep form of the model. The yielding process is described though not far as accurate as by the two-processes model. This especially counts for the fastest straining experiments. In fact the complex yielding behaviour found for HDPE can never be described accurately by one “Schapery creep or relaxation process”. Furthermore at small strains where yielding is absent the Schapery creep model gives better results than the Schapery relaxation model.
Figure 4.27 Stress during tensile straining experiments with strain rates of approximately 0.01 (○), 0.001 (△), . . . 0.000001 (○) 1/s and the Schapery relaxation model predictions (lines).

The predictions for ramp loading are given in figure 4.28. By comparison with figures 4.23 and 4.26 it can be concluded that all three models give equally good predictions in case of the ramp loading experiments. Since yielding did not play a role during these experiments; i.e., strain levels below 5%, this could be expected. The three predictions are also nearly identical at strain levels below 0.001. Therefore, it can be concluded that all three models give equal predictions for the linear viscoelastic region. Figure 4.21 shows that the Schapery relaxation model fit is linear for strain levels below 0.001.
Figure 4.28 Strain during ramp loading experiments with 4 (○), 8 (∇) and 12 (□) MPa reached in half an hour and the Schapery relaxation model predictions (lines).

4.3.2 Non-monotonous stress histories

In this paragraph stress histories with load reversal are considered. Three different test series were performed: recovery after creep, ramp unloading after ramp loading and stress relaxation. In the first series of experiments tensile bars were subjected to a constant engineering stress and unloaded after 19200 seconds of creep. Ramp loading was already discussed in section 4.3.1. The strains during unloading in the same manner after the load had reached its maximum are now investigated. Stress relaxation experiments were performed by application of a stepwise crosshead displacement which is held constant during relaxation. As a result the strain measured by the extensometer was constant within a few percent. Further details on the experimental set up are given in section 3.3.2.

The model predictions for recovery were evaluated similarly as for creep. Furthermore the numerical treatment of the calculations for ramp unloading is identical as for ramp loading. For the stress relaxation calculations a time decade was split in 32 steps equal on logarithmic scale. The solution of a time step is similar as for tensile straining. Again the strain measured with the extensometer was taken as input for the calculations.
Schapery creep model
The predictions of the Schapery creep model for recovery are given in figure 4.29. The experimental data are accurately described when the recovery times are smaller than 1000 seconds, meaning that superposition holds: \( \varepsilon(t_r) = \varepsilon_c(t_r + t_r) - \varepsilon_c(t_r) \). Only the recovery strain after creep at 12 MPa is underestimated over the entire region. This is the direct result of the inability to account for the yielding behaviour, which occurred before the recovery started. The creep model gives recovery curves which can be superimposed by vertical shifting on logarithmic strain scale. The measured recovery curves are also, although not exactly, superimposable in that manner.

At recovery times larger than 1000 seconds the recovery starts to decelerate more than expected from superposition. Eventually a plastic strain seems to emerge. Since no single plastic dashpot was used, viscoplasticity is not included in the model. The occurrence of a plastic strain is in agreement with creep recovery data of Zapas et al., [4.16].

**Figure 4.29** Strain recovery after creep experiments of 19200 seconds at load levels of 1 (○), 2 (▲), 4 (○), 6 (○), . . . 12 (○) MPa and the Schapery creep model predictions (solid lines), the Schapery viscoelastic viscoplastic model fit (dashed lines) and the plastic strains extrapolated from the measurements (●).
The recovery measurements of figure 4.29 indicate that after some recovery time a permanent plastic strain emerges. Recovery after creep at the same load levels but with shorter preceding creep times exhibit less or no plastic strains, see figure 4.36. This means that the plastic strain must increase with time during creep. Therefore the plastic strain is the result of viscoplasticity.

The plastic strain at the end of recovery can be estimated by extrapolation of the recovery curves, see figure 4.29. The plastic strain values, found by extrapolation, are shown as a function of the creep load level in figure 4.30. The values can be fitted with an Eyring dashpot, see section 2.2.3:

\[ \varepsilon_p = \dot{\varepsilon}_p t_c = \frac{\sigma}{\eta_b} \sinh \left( \frac{\sigma}{\sigma_0} t_c \right) \]

with \( t_c \) the creep time. The decreasing cross section and consequently increasing true stress during creep is not incorporated in this description of the plastic strain. This also explains the inaccuracy of the fit for 12 MPa, where the creep strain before recovery was ±15%.

**Figure 4.30** Viscoplastic strain found by extrapolation of the measured strain during recovery after 19200 seconds of creep as a function of the creep stress (crosses) and the Eyring fit (dashed line). (\( \sigma_0 = 2.9 \) MPa and \( \eta_b = 1.2 \times 10^8 \) MPa.s).

If the plastic strain of 0.0034 found after 19200 seconds of creep at 8 MPa engineering stress is the result of a constant viscoplastic strain rate, then this rate should equal \( 1.9 \times 10^7 \) s\(^{-1}\). However the creep strain rate at 19200 seconds is only slightly larger than this value and is decreasing far below this value for larger creep times. As a consequence the viscoplastic strain rate must be decreasing with the creep time. The Eyring dashpot therefore
cannot be valid, although the dependence of the plastic strain on the creep stress level is described properly by this model. Considering the two-processes model of section 4.2.3, the value of $\sigma_p = 2.9$ is very large compared with the values found in figure 4.10. See also the two-processes model of this section.

**Schapery creep model with viscoplasticity**

The implementation of viscoplasticity would be very straightforward if the creep response is not affected. In that case a distinction between viscoelasticity and viscoplasticity must appear only after unloading. As in [4.7, 4.16], the viscoplastic strain like the viscoelastic strain was assumed to be the product of a time and a stress function. In case of creep:

$$\varepsilon(t) = \varepsilon_0 + \varepsilon_{ve}(t) + \varepsilon_{vp}(t) = D_0 \sigma + \Delta D_{ve}(t) \sigma \bar{g}_2(\sigma) + \Delta D_{vp}(t) \sigma \bar{g}_3(\sigma)$$  \hspace{1cm} (4.14)

The creep behaviour of the original Schapery creep model fit is not altered by assuming that:

$$\Delta D_{ve}(t) = \Delta D_{vp}(t) = \Delta D(t) \hspace{1cm} \bar{g}_2(\sigma) + \bar{g}_3(\sigma) = g_2(\sigma)$$  \hspace{1cm} (4.15)

During unloading the viscoplastic strain is constant. The spring dashpot model of figure 4.31 was used for the interpretation of this viscoplastic strain and to enable the evaluation of the viscoplastic strain during arbitrary stress histories.

![Spring dashpot model of one Kelvin-Voigt element with viscoplasticity.](image)

*Figure 4.31*
The upper element in figure 4.31 is the viscoelastic element with its response evaluated with equation (4.3), but with $g_z(\sigma)$ replaced by $\bar{g}_z(\sigma)$. The element in the middle is the viscoplastic element active during tensile stresses whereas the lower element is active during compressive stresses. The extension element $k$ was evaluated with:

$$\dot{e}_q^{\text{ext}} = \dot{e}_k^{\text{ext}} + \max\left(0, D_k \bar{g}_3^{\text{vis}} \sigma_{n+1} - \dot{e}_k^{\text{ext}} \right) \frac{\Delta t_n}{\tau_k + \Delta t_n} \quad (4.16)$$

and the compression element $k$ with:

$$\dot{e}_q^{\text{comp}} = \dot{e}_k^{\text{comp}} + \min\left(0, D_k \bar{g}_3^{\text{vis}} \sigma_{n+1} - \dot{e}_k^{\text{comp}} \right) \frac{\Delta t_n}{\tau_k + \Delta t_n} \quad (4.17)$$

With the total strain equal to:

$$\varepsilon_{n+1} = D_0 \sigma_{n+1} + \sum_k (q_k^{\text{ext}} + q_k^{\text{comp}} + q_k^{\text{vis}}) \quad (4.18)$$

A disadvantage of this method is that the storage of all $q_k^*$ consumes three times more memory.

![Graph showing Leaderman (Schapery integral) function](image)

**Figure 4.32** Leaderman (Schapery integral) function of the original fit $g_z(\sigma)$ and those with the implementation of viscoplasticity $\bar{g}_z(\sigma)$ and $\bar{g}_3(\sigma)$.

The recovery curves of 1, 2, 4, 6 and 8 MPa were fitted with the Schapery creep model with viscoplasticity. No creep data were needed since the creep response of the model was not altered. The creep response up to 19200 seconds was evaluated similarly as was done in section 4.3.2. The strain recovery during zero stress was evaluated without
4.3 Arbitrary stress histories

Numerical treatments since the true stress is zero constantly. The viscoelastic true strain as a function of the recovery time was evaluated by:

\[ q_k(t_r) = q_k^0 \exp \left( -\frac{t_r}{\tau_k} \right) \]

with \( t_r \) the recovery time and \( q_k^0 \) the value of \( q_k^* \) at the end of the creep period. Equation (4.19) is exact since equation (2.67) is exact for constant stress levels. The Leaderman functions \( g_2(\sigma) \) and \( g_3(\sigma) \) were both set equal to a fourth order polynomial with their sum equal to \( g_2(\sigma) \), from section 4.3.2. The fitting was done similar as for the Schapery creep model.

![Graph](image)

**Figure 4.33** Strain during ramp unloading experiments with 4 (o), 8 (.) and 12 (□) MPa removed in half an hour after being applied similarly and the Schapery creep model predictions (solid lines) and the Schapery viscoelastic viscoplastic model predictions (dashed lines).

The results of the recovery fit are also shown in figure 4.29. Clearly the implementation of viscoplasticity with the additional fitting function \( g_3(\sigma) \) enables proper fitting of the recovery curves. Even the recovery curve found after creep at 10 MPa, which was not used during the fitting procedure, is described accurately. The recovery strain is slightly overestimated for the short creep times (100 - 10000 seconds) whereas the predictions from the Schapery model without viscoplasticity did not. From this it can be concluded that viscoplasticity occurs less at short creep times and more for longer times.
than expected from the assumption that the viscoelastic and viscoplastic spectrum are identical. If the viscoelastic spectrum and the viscoplastic spectrum were allowed to deviate from those found by the creep fit, then an almost exact recovery fit could have been achieved. The improvement would however be small while the fit would be more complicated.

The Leaderman functions $\bar{g}_3(\sigma)$ and $\bar{g}_5(\sigma)$ are shown together with the original function $g_2(\sigma)$ in figure 4.32. For numerical values see appendix A. The fraction of viscoplasticity, $\bar{g}_3/g_2$, turns out to increase slightly from 9% to 11% with the stress from 0 MPa to 8 MPa. At higher stress levels this factor increases to 18% for 12 MPa and 28% for 16 MPa. The latter value is the result of extrapolation of the fit to higher stress levels.

![Figure 4.34](image)

**Figure 4.34** Stress during stress relaxation experiments with strain levels of approximately 0.005 (○), 0.01 (▲), 0.02 (○), 0.04 (●), 0.08 (△) and 0.16 (◆) and the Schapery viscoelastic viscoplastic model predictions (dashed lines) and as comparison the Schapery creep model predictions (solid lines).
The two Schapery creep models were used to predict the material response during ramp unloading and stress relaxation. The results are given in figures 4.33 and 4.34, respectively. As can be observed the implementation of viscoplasticity improves the predictions for ramp unloading at longer unloading times. Apparently if unloading is to be predicted then the model also needs to be fitted to unloading cases. The restriction, that the model can not describe the yield behaviour and that consequently strains must be below 5%, still holds.

The role of viscoplasticity during stress relaxation is as follows. At short relaxation times the stress decreases due to the increasing viscoelastic strain in the fast Kelvin-Voigt elements. With increasing time also the slow elements start to stretch and the accompanying stress decrease results in a decreasing strain in the fast elements. The partial absence of contraction of the fast elements at large relaxation times because of plasticity results in a faster stress relaxation, since the plastic strain needs no stress to remain stretched.

![Graph showing stress relaxation](image)

**Figure 4.35** Comparison of full viscoelastic analyses (solid lines) and analyses using creep isochrones (dashed lines) with the Schapery creep model (no viscoplasticity) for stress relaxation with strain levels of 0.005, 0.01, 0.02 . . . . 0.16 (up in graph).

The differences between the Schapery model with and without viscoplasticity are negligible for stress relaxation at strain levels of approximately 1 and 2%, see figure 4.34. Hence it can be concluded that viscoplasticity does not play an important role for stress
relaxation at these strain levels and relaxation times. At larger strain (stress) levels the fraction of viscoplasticity $\bar{g}_3/g_2$ increases and also the time dependent strain with its viscoplastic part increases relative to the linear elastic strain. Apparently the overestimation of the relaxation stress at larger strain levels with the Schapery creep model can, besides by the inability to describe yielding, also be caused by the absence of viscoplasticity. One has to bear in mind that the large value of $\bar{g}_3/g_2$ at high stress levels, see figure 4.32, was obtained by fitting plastic strain due to low creep stress levels. The results in figure 4.34 indicate that the fraction of viscoplasticity is overestimated at large stress levels, since the stress during stress relaxation at 4% strain is underestimated when viscoplasticity is included.

![Graph showing strain recovery](image)

**Figure 4.36** Strain recovery after creep experiments at a load level of 8 MPa with creep times of 150 (.), 300 (.), 600 (o), 1200 (.), . . . 19200 (.) seconds and the Schapery creep model predictions (solid lines) and the Schapery viscoelastic viscoplastic model predictions (dashed lines).
In section 4.3.1 the (full viscoelastic) response of the Schapery creep model was compared with analyses using the creep isochrones. Now both analysis are repeated for stress relaxation. The results are shown in figure 4.35. The differences are less than 5%. It can therefore be concluded that in our case the creep modulus $1/D(t)$ is a good approximation of the relaxation modulus $E(t)$. Also the stress/strain nonlinearity is described properly. Only at high strain levels the approximation fails due to the onset of yielding. The results are in agreement with those found by Struik [4.17] and Ferry [4.18].

**Figure 4.37** Strain during multiple creep recovery experiments at load levels of 4 (○), 8 (△) and 12 (□) MPa and the Schapery viscoelastic viscoplastic model predictions (dashed lines). The creep times were 150, 300, 600, 1200 and 2400 seconds. The recovery times were 5 times longer than the preceding creep time (except last cycle).
Figure 4.36 shows the measured recovery strain after creep at 8 MPa with varying creep times. As can be observed the implementation of viscoplasticity improves the description of recovery. However the creep data at short times were described better if no viscoplasticity was used. One can again conclude from this that the viscoplastic strain at short times is smaller than found with the assumption that the viscoplastic spectrum equals the viscoelastic spectrum.

The predictions and measurements for multiple creep recovery are shown in figure 4.37. Again the relative deviations from the measurements are within 10 to 20%. Possible errors do not increase with the stress cycle applied.

**Figure 4.38** Strain recovery after creep experiments of 19200 seconds at load levels of 1 (•), 2 (▼), 4 (○), 6 (●) . . . 12 (□) MPa and the two-processes model predictions; fit 1 (solid lines) and fit 2 (dashed lines).

**Two-processes model**

The parameters of the two-processes model found by fit 1 of the creep data, see the solid lines in figures 4.11 and 4.12, were used to predict recovery data. The results are given in
4.3 Arbitrary stress histories

figure 4.38 as solid lines. As can be observed the data are not predicted properly. The recovery curves predicted by the two-processes model seem to superimpose by shifting horizontally + vertically instead of vertically as measured.

The two-processes model does contain a plastic dashpot in process 1 though process 2 does not. As a result all strains in the material will disappear eventually according to this model. However one can imagine that this model shows the following recovery. At creep times of 19200 seconds the stress in process 1 has vanished, see figure 4.15, and all the creep strain in process 1 is concentrated in its plastic dashpot, while the creep stress is carried completely by process 2. After sudden unloading process 1 will be loaded with $-\frac{1}{2}\sigma$ and process 2 with $\frac{1}{2}\sigma$ where $\sigma$ is the creep load. Since at small creep times both processes carried loads of approximately $\frac{1}{2}\sigma$ the initial stages of recovery predicted by this two process model will be close to that predicted by superposition, i.e., the Leaderman creep model. After some recovery time the stresses in both processes drop and the recovery rate decreases very fast due to the freezing in of the strains, i.e., time-stress superposition increases the retardation times of both processes. Only after very long recovery times, which were not measured, complete recovery occurs.

![Graph of creep strain vs creep time](image)

**Figure 4.39** Creep strain at load levels of 1, 2, 3, ..., 10 MPa (up in graph) of a hypothetical viscoelastic material obeying time-stress superposition with Eyring parameter $\sigma_0 = 0.3$ MPa.
To understand the failure of the two-processes model during recovery a creep recovery simulation was made with a hypothetical material obeying time-stress superposition (one process). The material has a power law spectrum and a single dashpot for yielding. Furthermore the Eyring function, see equation (4.6), was taken for time-stress superposition and the Eyring parameter $\sigma_0$ was set equal to 0.3 MPa. Its creep response is shown in figure 4.39.

![Graph showing recovery strain at 1000 seconds of creep at load levels of 1, 2, 3... 7 MPa (up in graph) of a hypothetical viscoelastic material obeying time-stress superposition with Eyring parameter $\sigma_0 = 0.3$ MPa.]

**Figure 4.40** Recovery strain at 1000 seconds of creep at load levels of 1, 2, 3... 7 MPa (up in graph) of a hypothetical viscoelastic material obeying time-stress superposition with Eyring parameter $\sigma_0 = 0.3$ MPa.

The recovery strain after 1000 seconds of creep is shown in figure 4.40. As can be observed the recovery curves superimpose by shifting horizontally + vertically. This was also the case for the predicted recovery curves of figure 4.38. Apparently the recovery predicted by the two-processes model is mainly determined by process 2. The viscoelastic strain after 100 seconds of recovery is plotted as a function of the creep load in figure 4.41. Except for recovery after creep at 1 MPa, where the strain already starts to recover, the strain values can be fitted with an Eyring dashpot, see equation (4.13). However the Eyring parameter $\sigma_0$ of the fit equals 0.917 MPa and not 0.3 MPa. The dependence of the recovery strain on the creep stress is not only determined by the Eyring parameter of time-stress...
superposition but also by the slope of the power law creep curve.

Assuming that the apparent viscoplastic strain found after recovery is actually the viscoelastic strain in process 2 then the Eyring parameter of figure 4.30 is not the Eyring parameter of the time-stress superposition of process 2. The recovery of this viscoelastic strain is very slow due to its “freezing-in” by time-stress superposition. The result is an apparent viscoplastic strain.

![Graph showing creep strain vs. creep stress](image)

**Figure 4.41** Recovery strain of a hypothetical viscoelastic material at 100 seconds of recovery after 1000 seconds of creep as a function of the creep load level together with an Eyring dashpot fit with $\sigma_0 = 0.971$ MPa.

The recovery time at which the recovery curve in figure 4.40 bends downward depends on the effective time reached just before the start of recovery and therefore, besides the creep time, solely depends on the Eyring parameter of the time-stress superposition. The recovery time at the bend in figure 4.29 is too short compared with the data. As a result the Eyring parameter of process 2 must be smaller than the value of 0.6 found in figure 4.14. Considering the bends of figure 4.40 the parameter must also be smaller than the value used in the simulation, i.e. 0.3. For figure 4.10 this implies that the dashed line of process 2 must have a smaller slope and that the curvature found in the data continues slightly for smaller strain rates.

Bearing the experience with the hypothetical material in mind, the creep curves were fitted for the second time. Both processes obey time-stress superposition with the Eyring function. The Eyring parameter of process 2 was set equal to 0.1 MPa as starting value. Both Eyring values, elastic compliances and spectra were used to fit the creep data. The fit 2 is given in
figure 4.42 as dashed lines. The spectra and other material parameters are given in figure 4.43. As can be observed in figure 4.43, the spectra were built up with pieces of straight lines. The Eyring parameters are: $\sigma_0^1 = 1.98$ MPa and $\sigma_0^2 = 0.0984$ MPa. The values of $a_\sigma^2$ can become very large: $10^{40}$ when the stress in process 2 is 10 MPa.

The Eyring parameter of process 2 hardly changed in value during the fitting of the creep data. Increasing or decreasing its starting value did not result in better fits of the creep data. In figure 4.42 it can be observed that the main deviation from the measurements is the result of the gradual convex curvature which could not be described by the model. At large creep times, when the stress in process 1 has almost vanished, the creep strain rate is much lower than those of the measurements. This is caused by the low value of $\sigma_0^1$. If the creep stress is increased the accompanying large increment of $a_\sigma^2$ will not result in a too large increase of the creep strain of process 2 when the creep curve is very flat.

**Figure 4.42** Strain during creep experiments at load levels of 1 (v), 2 (v), 4 (o), 6 (o) ... 18 (o) MPa and the two-processes model fit 2 (dashed lines).
4.3 Arbitrary stress histories

If process 2 indeed has a very flat creep compliance curve, then the larger slope of the creep curves at creep times from 10000 seconds until the end of the data must be the result of the stress transfer from process 1 to process 2. Also the recovery after 19200 seconds of creep can then be explained. The larger amount of the creep stress is still carried by the slightly nonlinear process 1. This process will exhibit full recovery, while process 2 will cause the apparent plastic strain. In fact the plastic strain is the very slowly recovering viscoelastic strain in process 2.

Unfortunately a good fit of the creep curves with the two-processes model with Eyring-like time-stress superposition could not be obtained. The second fit however does exhibit a higher stress in process 1 than fit one, compare figures 4.44 and 4.15. The recovery behaviour of the fit 2 is given in figure 4.38 as dashed lines. Although the data are still not correctly predicted quantitatively, the recovery predictions have improved qualitatively compared with those of fit 1. The predicted curves of 4, 6, 8 and 10 MPa can be almost superimposed by vertical shifting and also after some recovery a plastic strain emerges. This plastic strain is however far too large in value. The predictions for tensile straining and ramp loading are given in figures 4.25 and 4.26 as dashed lines. Also for fit 2 the predictions are accurate.

Figure 4.43 Retardation spectra (Kelvin-Voigt parameters) of the two-processes model fit 2. \( (D_0^1 = 0.000821 \text{ } \text{ } 1/\text{MPa}, \text{ } D_0^2 = 0.00187 \text{ } \text{ } 1/\text{MPa}, \text{ } \eta_1 = 3.24 \cdot 10^7 \text{ } \text{ } \text{MPa.s} \text{ } \text{ } \text{and} \text{ } \eta_2 = 3.07 \cdot 10^{58} \text{ } \text{ } \text{MPa.s}) \)
Figure 4.44 True stress in process 1 during creep at 1, 2, 4, 6, 8, 10, 12, 14 MPa for fit 2 (up in graph).

Figure 4.45 True stress in process 2 during creep at 1, 2, 4, 6, 8, 10, 14 MPa for fit 2 (up in graph).

Figure 4.46 Strain during ramp unloading experiments with 4 (o), 8 (Δ) and 12 (□) MPa removed in half an hour after being applied similarly and the two-processes model prediction using fit 1 (solid lines) and fit 2 (dashed lines).

The predictions for ramp unloading are in good agreement with the measurements for both fits, see figure 4.46. The curve at the highest stress recovers slightly more than predicted with the model. As for recovery the unloading is too slow. The stress relaxation predictions of the two process model are shown together with the measurements in figure
4.47. The model with the parameters of the fit 1 gives good predictions if the strain level remains below 2%. As for creep fit 2 with the two process model gives a too sharp (convex) curvature. At higher strain levels the stress is overestimated.

![Stress relaxation experiment](image)

**Figure 4.47** Stress during stress relaxation experiments with strain levels of approximately 0.005 (∇), 0.01 (♦), 0.02 (○), 0.04 (▪), 0.08 (△) and 0.16 (▲) and the two-processes model predictions using fit 1 (solid lines) and fit 2 (dashed lines).

In case of the Schapery creep model this overestimation of the stress during stress relaxation at high strain levels could be attributed to the inability to describe yielding and the absence of viscoplasticity. For fit 2 of the two-processes model the apparent viscoplastic strain after creep and recovery is overestimated and yielding during tensile straining is described properly. A specific cause of the deviations at high strain levels is therefore difficult to give.

The inability of the model to fit the creep and recovery curves of HDPE can have many causes. As was stated in section 4.2.2, the difference in stiffness of shell and core due to the injection moulding process, see section 3.2 and figure 5.14, can cause different creep
spectra and nonlinearities in the shell and core. This may cause the convex curvature of the creep curves to appear at different creep times for the shell and core layer. The result of this will be a more gradual convex curvature than expected from the two-processes model.

Struik [4.17] pointed out that merely the composite structure of the crystalline and amorphous regions will result in a multi-phase amorphous structure. Segmental motions in the amorphous regions close to the crystals will be restricted by the crystals whereas regions far from the crystals will not suffer from these restrictions.

In section 4.6 it is concluded that process 1 describes the crystalline phase and process 2 probably describes the amorphous phase. However dynamic mechanical analyses have shown that for HDPE two processes are responsible for the mechanical behaviour of the crystalline phase, [4.18]. Therefore the discrepancy between data and two-processes model predictions could also be caused by the presence of three processes in HDPE: two slightly nonlinear processes (1 and 1') for the crystalline phase and one highly nonlinear process (2) for the amorphous phase. By taking two almost linear processes instead of one, the convex curvature during creep can be described by 1 and 1', leaving the parameters of process 2 free to fit the apparent plastic strain after creep recovery.

The low value of $a_\sigma^2$ results in a very long retardation spectrum with many Kelvin-Voigt elements for process 2. The large number of elements requires a large number of $q_k$ to be stored into computer memory, see section 2.3.1. If the two-processes model is ever to be successful in numerical applications such as FEM then a faster numerical treatment is needed. In case of time-stress superposition with very small values of $\sigma_0$ and consequently very large values of $a_\sigma$ the process response can be approximated with:

$$\varepsilon(t) = D(\psi_{\text{max}}')\sigma_{\text{max}} - D(\psi_{\text{max}}' - \psi'(t))(\sigma_{\text{max}} - \sigma(t))$$

(4.20)

with a modified effective time:

$$\psi'(t) = \int_0^t \text{sign}(\sigma(\xi) - \sigma_{\text{max}}) \frac{d\xi}{a_\sigma(\xi)}$$

(4.21)

where $\sigma_{\text{max}}$ is the highest stress value reached during the loading history, $\psi_{\text{max}}'$ the largest value of the modified effective time and sign($x$) equals 1 for $x \geq 0$ and equals -1 for $x < 0$. Note that this approximation is only valid for tensile stresses and when unloading results in the "freezing-in" of strains with large retardation times which will only "unfreeze" due to application of stresses higher than $\sigma_{\text{max}}$. In case of creep recovery the plastic strain approximately becomes $(D(\psi_{\text{max}}') - D(0))\sigma_{\text{max}}$.

This approximation also explains why changing $a_\sigma^2$ hardly affected the fit of the
creep curves. In case of creep the stress in process 2 is increasing. Therefore its strain becomes:

\[ \varepsilon(t) = D(\psi_{\text{max}}')\sigma_{\text{max}} \]

(4.22)

A change of \( a_2^p \) results in a change of \( \psi_{\text{max}}' \) which can be compensated by changing the creep compliance function.

**Figure 4.48** Strain recovery after creep experiments of 19200 seconds at load levels of 1 (△), 2 (○), 4 (●), 6 (●), ... 12 (□) MPa and the Schapery relaxation model predictions (lines).

**Schapery relaxation model**

This model failed as well in predicting the recovery behaviour because of absence of viscoelasticity. Furthermore this model does not exhibit recovery curves which can be superimposed by vertical shifting on logarithmic strain scale since the nonlinearity is caused by the strain. In figure 4.48 it can be observed that indeed predictions for recovery are poor.
Even for small recovery times, where the role of the plastic strain is nearly negligible, the model fails. Therefore no attempts were made to include viscoplasticity in the relaxation model. Also the mechanical analogy of viscoplasticity for the relaxation model will be more difficult than for the creep model. The latter was given in figure 4.31.

The strain data of ramp unloading are given together with the Schapery relaxation model predictions in figure 4.49. Clearly the model fails when the load level reaches values close to zero. This inadequacy is directly related to the inadequacy to describe the viscoplastic strain emerging during recovery.

![Graph showing strain versus unloading time](image)

**Figure 4.49** Strain during ramp unloading experiments with 4 (○), 8 (♂) and 12 (□) MPa removed in half an hour after being applied similarly and the Schapery relaxation model predictions (lines).

The Leaderman relaxation model, obtained from the Schapery relaxation model by $g_0 = g_1 = a_0 = 1$ ($E_0 << E_1$), is based on the assumption that stress relaxation curves can be superimposed by vertical shifting on logarithmic stress scale. As can be observed in figure 4.50 this is indeed the case. The experimental results are therefore in agreement with those found by Popelar et al. [4.5]. And although the Leaderman relaxation model is based on this superimposability of the stress relaxation curves, when fitted to creep data, it does not provide better predictions for stress relaxation than the Leaderman (Schapery) creep model.

The Schapery relaxation model qualitatively yields the same predictions for stress relaxation as the Schapery creep model. During the fitting of the creep curves it was found that the linear spring necessary for the Schapery relaxation model to fit the creep data almost
vanished for the relaxation model, see section 4.2.3. This implies that although the creep curves do not superimpose completely by a vertical shift, the relaxation curves do, indicating that separation of variables for stress relaxation holds, see equations (4.1, 4.8).

The stresses found during relaxation at strain levels of 8% and 16% differ only a few percent indicating that yielding has occurred. The yield behaviour found during creep could be described reasonably well with this model, however the relaxation data at large strain levels are overestimated. This can, as for the creep model, be the result of the absence of viscoplasticity or of the overestimation of the yielding stiffness at high stress levels.

**Figure 4.50** Stress during stress relaxation experiments with strain levels of approximately 0.005 (○), 0.01 (▲), 0.02 (●), 0.04 (●), 0.08 (○) and 0.16 (▲) and the Schapery relaxation model predictions (lines).

### 4.4 The effect of temperature on creep

Creep experiments were performed 64 weeks after production at various temperatures. The temperatures ranged from 18 to 43°C with temperature steps of 5°C. Figure 4.51 shows the
creep strain measured at these temperature levels and a load level of 10 MPa. As can be seen the curves can impossibly be shifted upon a master curve. The shape of the creep curve at 18\degree C is different from that at 43\degree C. At 18\degree C the convex curvature is present whereas at 43\degree C the curve has become concave over the entire time region. It can therefore be concluded that the creep behaviour is thermo-rheologically complex, see section 2.2.4.

![Graph showing creep strain vs creep time](image)

**Figure 4.51** Strain during creep experiments at a load level of 10 MPa and temperatures of 18 (\text{\textdegree}C), 23 (\text{\textdegree}C), 28 (\text{\textdegree}C), \ldots, 43 (\text{\textdegree}C) \text{\degree}C.

The effect of temperature on creep seems qualitatively equivalent to the effect of stress. However the description of the creep curves for all stress levels needed two parallel processes with different time-stress superposition functions. The thermo-rheological complexity can therefore be explained by the difference in the time-temperature superposition functions of these two processes, [4.14].
4.4 The effect of temperature on creep

Schapery creep model
The creep behaviour of HDPE below strain levels of 5% could be described by the Schapery creep model. This model has only one process which contributes to the creep strain. Likewise creep measurements at different temperatures indicate that the creep strains below 5% can be described by a thermo-rheologically simple material model, i.e., superposition by horizontal and vertical shifting of a master curve, see figures 4.52 - 4.54.

![Image of a graph showing creep strain vs. creep time with different symbols for different temperatures.]

Figure 4.52 Strain during creep experiments at a load level of 2 MPa and temperatures of 18 (o), 23 (v), 28 (o) . . . 43 (a) ºC and the fit with the Schapery creep model with temperature shifting (solid lines).

Therefore the creep data measured at 18, 23, 28 . . . 43ºC with strains below 5% were fitted with the Schapery creep model of section 4.2.1. The material parameters found in section 4.2.1 were not altered and the temperature effect was assumed to obey equation (2.38). The creep strain at a constant true stress then becomes:

\[
\varepsilon_c(t) = \left( D_0 \sigma + \Delta D(t/a_p) \sigma g_2(\sigma) \right) g_1
\]

(4.23)
The creep curves should therefore be superimposable on double logarithmic scale.

Since the creep experiments were performed 64 weeks after production and the Schapery model was fitted with data obtained 32 weeks after production, a vertical \( g_r \) and horizontal shift \( a_r \) were also allowed for 23°C. This would then compensate small ageing effects, see section 4.5. No prescribed functions were taken for the shifting functions \( g_r \) and \( a_r \), their values for the creep temperatures were used to fit the creep data. The fitting was done with the same method as with the Schapery creep model.

![Figure 4.53](image)

**Figure 4.53** Strain during creep experiments at a load level of 4 MPa and temperatures of 18 (○), 23 (▴), 28 (○) . . . 43 (▲) °C and the fit with the Schapery creep model with temperature shifting (solid lines).

The results of the fitting procedure are given in figures 4.52 - 4.54. As can be observed the data are described properly up to 5% strain. Above this strain level yielding commences. One can conclude from this that although HDPE is a thermo-rheologically
complex material, the effect of temperature on creep can be described by a thermo-
rheologically simple model as long as yielding is absent.

The horizontal and vertical shift values are shown in figures 4.55 and 4.56,
respectively as a function of the reciprocal temperature in Kelvin. In both figures the
shifting values are situated on a straight line. The shifting values can therefore be described
by the Arrhenius equation (2.39). The straight solid lines in figures 4.55 and 4.56 denote the
best fit of the shifting values with this equation. However since the temperature range is
rather small the values could be described by the WLF equation as well, see equation (2.42).
The WLF fits are given in the figures as dashed lines. As can be observed these lines do not
yield a straight line in an Arrhenius plot.

![Graph showing strain during creep experiments at a load level of 8 MPa and
temperatures of 18 (•), 23 (○), 28 (○) . . . 43 (△) °C and the fit with the Schapery
creep model with temperature shifting (solid lines).]
Figure 4.55  Arrhenius plot of the horizontal shifting values found by fitting of creep data as a function of the reciprocal creep temperature (diamonds), the Eyring fit (solid line, $\Delta H = 4.00 \times 10^4$ J/mole, $T_0 = 295$ K) and the WLF fit (dashed line, $C_1 = 4.83$ K, $C_2 = 76.4$ K, $T_0 = 295$ K).

Figure 4.56  Arrhenius plot of the vertical shifting values found by fitting of creep data as a function of the reciprocal creep temperature (diamonds), the Eyring fit (solid line, $\Delta H = (-) 7.05 \times 10^3$ J/mole, $T_0 = 298$ K) and the WLF fit (dashed line, $C_1 = (-) 194$, $C_2 = 2.10 \times 10^4$ K, $T_0 = 298$ K).
4.4 The effect of temperature on creep

The vertical shifting values shown in figure 4.56 are rather large. Vertical shifting values mostly have been attributed to the density changes caused by the temperature variation, see section 2.2.4, and should therefore be much smaller. However as was pointed out by Klompen et al. [4.14], the horizontal shifts of the two parallel processes caused by a temperature increase can result in a vertical shift of the creep curve, just as was the case for horizontal shifts caused by a stress increase. The large vertical shifting values can be explained by the model with two parallel processes. The numerical parameters of the Arrhenius and the WLF equation ($\Delta H$, $C_1$ and $C_2$) have therefore no physical meaning for vertical shifting. However, since the position of the convex curvature in the creep curves is related to the end of the spectrum of process 1 of the two-processes model, the horizontal shifting values do have a physical meaning; they represent the time-temperature superposition of this process 1.

Two-processes model
Figure 4.51 clearly showed that HDPE exhibits thermo-rheologically complex creep behaviour. The effect of temperature on creep is qualitatively similar to the effect of stress on creep. The yield behaviour, see figure 4.10, and also the creep behaviour of HDPE requires at least two parallel processes to be described over the entire strain range measured, see section 4.2.2. Each process obeys time-stress superposition. Since the effect of stress and temperature on creep are similar, it is very likely that the effect of temperature can be modelled with two parallel processes with each obeying time-temperature superposition.

Numerical simulations for a thermo-rheologically complex hypothetical polymer were performed by Klompen et al. [4.14]. Their simulation showed that indeed large vertical shifts are obtained due to the time-temperature superposition (horizontal shifting) of each process. The temperature independence of the strain level above which the Schapery creep model is no longer valid due to the start of yielding, i.e., 5%, indicates that additional vertical temperature induced shifts, $g_1^2$ and $g_2^2$, are probably very small. The horizontal temperature shifts found with the Schapery creep model can be attributed to the time-temperature superposition of process 1, see end of Schapery creep model in section 4.4.

Since our fit of HDPE was only accurate when irregular time-stress superposition functions were used, see figure 4.14, an attempt to fit the creep data over the entire strain region for all temperature levels was not made.

Schapery relaxation model
The fit of the creep data with the Schapery relaxation model is good over the entire strain region, see figure 4.17. Implementation of the principle of time-temperature superposition in this model will only result in horizontal shifts of the creep curves when creep occurs at various temperatures. Consequently large vertical shifts are required to fit the creep data of,
for instance, figure 4.52. Since this model is a relaxation model, with the nonlinearity described by a strain function $h_2(\varepsilon)$, this vertical shifting must be implemented at this strain function. In equation (2.19) $H(\varepsilon)$ must be replaced by $H(\varepsilon)h_p(T)$.

Because of $h_p(T)$ the same creep curve can be obtained at a different temperature only with a different stress level. As was stated earlier the effect of temperature on creep is similar to the effect of stress. Consequently the Schapery relaxation model is capable to at least qualitatively describe the complex behaviour of figure 4.51. The horizontal shift values of the Schapery creep model must also be found with this relaxation model. However this model was not used to fit the creep curves at all temperatures.

4.5 The effect of ageing on creep

Creep experiments at 23°C were performed 2, 4, 8 . . . . 128 weeks after production on virgin state specimens. Since the creep time was only 2 hours, the effect of ageing during the creep experiment itself can be neglected, [4.18]. The experiments were performed at load levels of 2, 4, 8 and 12 MPa at 23°C. The results of 4, 8 and 12 MPa are given in figures 4.57 - 4.59. Since the experimental errors were too large compared with the ageing effect for the 2 MPa creep curves, the analysis of ageing was restricted to load levels of 4, 8 and 12 MPa.

The effect of ageing in HDPE has been described by Struik [4.4, 4.19] and Lai et al. [4.6]. Both used the time-ageing time superposition, i.e., horizontal shifting on logarithmic time scale, together with vertical shifting on linear strain scale. In case of creep for a single stress level this leads to:

$$\varepsilon(t) = D_0 g_{oe} + AD(t/a_e)$$  \hspace{1cm} (4.24)

The vertical shifting accounts for changes in the elastic compliance and has no effect on the strain rate during creep. Therefore the creep strain rates for all tested material ages were fitted using a creep spectrum and shifting values $a_e$ for each stress level to minimize the relative quadratic errors, see also the fitting with the Schapery creep model. Two Kelvin-Voigt elements per decade were taken for the creep spectra for extra smoothness of the strain rate curves. The effect of $g_{oe}$ is considered so small that differences in true stress do not change the creep curves with ageing time.

The shifting values $a_e$ resulting from the fitting procedure are shown in figure 4.60. If the same method as Struik and Lai is used then the aging rate $\mu$ is a function of the creep stress level. Although the strain curves for 8 and especially 4 MPa do not exhibit a gradual change with ageing time the fitting results on the strain rate are regular. The scatter in the
strain data are therefore considered to be the result of scatter in the zero strain reference, see section 3.5. Ageing experiments by Struik and Lai were performed on a single specimen, which was given a cyclic load. As a result the zero strain error will be equal for all creep curves. Furthermore Lai experimented with quenched specimens leading to more ageing, \( \mu = 0.69 \), see figure 4.60 for our values.

![Graph showing creep strain over time with different markers for different ages.](image)

**Figure 4.57** Strain during creep experiments at a load level of 4 MPa performed 2 (\(\circ\)), 4 (\(\triangledown\)), 8 (\(\diamond\)) . . . . 128 (\(\vartriangle\)) weeks after injection moulding.

An ageing rate which changes with the stress level would imply that the ability to superimpose strain rate curves vertically on logarithmic scale is only valid at a certain age and that its successful use in our case was a matter of sheer luck. At other material ages the difference in ageing rates will cause horizontal shifts of the creep curves on logarithmic time scale which depend upon the stress level. In case the complete creep curves shift with ageing, then the pronounced convex curvature will occur at different creep times and consequently invalidate the superimposability.
Figure 4.58  Strain during creep experiments at a load level of 8 MPa performed 2 (v), 4 (w), 8 (o) . . . 128 (□) weeks after injection moulding.

In case of vertical shifting of the creep curves on logarithmic strain scale instead of linear strain scale, a vertical shift would also modify the creep strain rate. Vertical shifting on logarithmic strain scale has been performed for other polymers, [4.19]. In case of creep at a given stress this leads to:

$$\varepsilon_c(t) = \left(D_0 + \Delta D(t/a_e)\right) g_e$$  \hspace{2cm} (4.25)

This means that the effect of ageing and temperature are modelled equivalently, compare equation (4.23). The strain rate data were fitted with values of $a_e$, which are independent of the creep stress level, and a shifting angle which is independent of the stress level and age.
4.5 The effect of ageing on creep

Figure 4.59 Strain during creep experiments at a load level of 12 MPa performed 2 (○), 4 (▼), 8 (○) . . . . 128 (○) weeks after injection moulding.

Figure 4.60 Shifting values $a_e$ as a function of the ageing time for the creep strain rate fits for the stress levels of 4 (○), 8 (△) and 12 (□) MPa and their fits with a constant ageing rate (solid lines; 4 MPa: $\mu = 0.255$, 8 MPa: $\mu = 0.142$ and 12 MPa: $\mu = 0.0930$).
The difference in the ageing rates can then be explained by the difference in the angle of the creep curves on double logarithmic scale, see figure 4.61. A horizontal shift of a decade of a creep curve results in a horizontal shift and a vertical shift of a decade of the creep strain rate. The horizontal shifting values $a_e$ which depend on the stress level might be replaced by shifts independent of the stress level but with an angle to the original shifts. The apparent dependence of $a_e$ on the creep stress level which was found earlier, is then caused by the slight differences in the slope of the creep curves. Furthermore the ageing creep data have a time range of 10 to 7200 seconds, which is the region where power law approximation of the creep curve is allowed. Therefore a clear distinction between horizontal and horizontal + vertical shifting on double logarithmic scale cannot be made on basis of these data.

A new fit of the strain rate was made. As already described, the spectra, the shifting angle and the shifting values $a_e$ were varied until the sum of the errors of all three stress levels reached its minimum. The shifting values $a_e$ and the spectra can be found in figures 4.62 and 4.63. The constant shifting angle determined by the fit was:

$$\log(g_e) = -0.432 \log(a_e)$$

The average relative quadratic errors of the strain rate were: 0.010, 0.0013 and 0.00097 for 4, 8 and 12 MPa, respectively. These values are in the order as for the first fit with horizontal shifting and different ageing rates: 0.012, 0.0023 and 0.00066. As expected the three spectra exhibit different slopes, see the solid lines in figure 4.63. This implies that the vertical superimposability of the strain rate curves is not exactly valid, though a good approximation.
4.5  The effect of ageing on creep

![Graph showing the relationship between ageing time and log(a_e)](image)

**Figure 4.62**  Shifting values $a_e$ as a function of the ageing time for the creep strain rate fit and its fit with a constant ageing rate (solid line: $\mu = 0.0436$).

After the transient creep compliances $\Delta D(t)$ had been determined for the stress levels 4, 8 and 12 MPa, the creep strain data were fitted with the elastic compliances $D_0$. The fits are shown as solid lines in figures 4.57 - 4.59. If the proposed model for ageing is correct and the fit of the strain rates is very accurate, there should be no relation between the ageing time and the elastic compliances. The values found by the fit are plotted in figure 4.64 as a function of the ageing time.

![Graph showing the retardation spectra for different stress levels](image)

**Figure 4.63**  The retardation spectra for the 4 (●), 8 (△) and 12 (□) MPa creep strain rate fit and their power law fits for the smooth center parts (solid lines).
Since the zero strain error is relatively smallest for the 12 MPa creep curves the results for this stress level are most significant. The 12 MPa data in figure 4.64 show that the fit gives an increase of the elastic creep compliance with the ageing time, which is opposite to what is expected. The irregularity of the data in figure 4.64 is caused by the fact that the strain error and the effect of ageing are in the same order of magnitude. Therefore further modelling of ageing was not done.

Furthermore the effect of ageing for larger creep times might be more complicated due to the presence of two viscoelastic processes, which in principle could have different ageing characteristics. Like the effect of temperature, the effect of ageing can result in two different $a_o$ for the two processes and result in a vertical shift of the creep curve on logarithmic strain scale. Also the injection moulding process can cause different ageing characteristics of the skin and core layer, see section 3.2 and figure 5.13.

![Graph](image)

**Figure 4.64** Ratio of the elastic compliances for the 4 (○), 8 (△) and 12 (□) MPa creep strain fits; 4 MPa: $D_0 = 0.000518$ 1/MPa, 8 MPa: $D_0 = 0.000544$ 1/MPa and 12 MPa: $D_0 = 0.000749$ 1/MPa.

### 4.6 Creep of different grades of HDPE

The creep response of three different grades of HDPE was measured 32 weeks after production at a temperature of 23°C. For further details see section 3.3.5. The results of creep experiments on HDPE 7058 (DSM) were discussed in sections 4.2.1 to 4.2.3 and are shown in figure 4.1. The second and third test series were performed on HDPE 7108 (DSM) and HDPE Rigidex HM5420XP (BP Chemicals).

HDPE 7108 like 7058 is an injection moulding grade but has a lower molecular
4.6 Creep of different grades of HDPE

weight resulting in a higher melt index, see table 3.1. The creep data together with the experimental results of 7058 can be found in figure 4.65. The results of the extruded pipes of Rigidex HMS420XP are plotted together with the experimental results of 7058 in figure 4.66. As can be observed all three grades exhibit the same qualitative creep behaviour and therefore the applicability of the viscoelastic models is identical for all grades.

The results of figure 4.65 clearly show that differences in the creep behaviour of HDPE 7058 and 7108 are minimal. Apparently the difference in MFI, $M_s$ or $M_w$ do not necessarily determine the viscoelastic behaviour of HDPE. Note that the injection moulding conditions were similar. The differences between HDPE 7058 and our Rigidex grade are more apparent as can be seen in figure 4.66. Despite the less accurate data it can be concluded that the Rigidex exhibits stiffer behaviour at high stress levels and short creep times. However at large creep times the material exhibits more creep strain. The time needed for yielding at 10 MPa will be larger since the Rigidex curve is still convex for $10^6$ seconds.

![Diagram](image)

**Figure 4.65** Strain of HDPE 7108 during creep experiments at load levels of 1 (v), 2 (v), 4 (o) ... 18 (♦) together with smooth fits of the HDPE 7058 data (solid lines).
Figure 4.66  Strain of HDPE HM5420XP during creep experiments at load levels of 1 (v), 2 (v), 4 (o) . . . . 18 (•) together with smooth fits of the HDPE 7058 data (solid lines).

Three-point bending dynamic mechanical experiments on prismatic beams of the three HDPE grades were also performed. The beams measured 3.6 mm in width and ±1.8 mm in thickness and the length between the outer supports was 20 mm, see also section 3.3.5. The complex modulus $E^*$ and the phase lag $\delta$ measured at a frequency of 1 Hz are shown in figure 4.67 as a function of the temperature.

The imposed force amplitude can introduce maximum stresses large enough to cause material nonlinearities. This together with the variations in thickness of the specimens will introduce quantitative differences in the measured complex modulus. The phase lag however can be regarded as a material property which allows qualitative comparison.

The glass transition temperature revealing itself in the first peak in the phase lag of figure 4.67 is situated at -115°C for all three materials. The phase lag of HDPE 7058 and 7108 are nearly identical, especially from 0°C onwards. From figure 4.65 it was found that their creep behaviour was also nearly identical. The extruded Rigidex HM5420XP exhibited a different creep response and in figure 4.67 it can be observed that the phase lag exhibits a
significant difference with the other two as well. The second upward part, corresponding to the crystalline phase, starts at a higher temperature. From this observation it can be concluded that the crystallization temperature of Rigidex is higher than those of 7058 and 7108. This is in agreement with given crystalline volume fractions in table 3.1.

The stiffer mechanical behaviour at short creep times of Rigidex can be attributed to the larger crystalline fraction. In terms of the two-processes model of section 4.2.3 and 4.3.2, this means that process 1 can be attributed to the crystalline phase. Process 2 is then probably the result of the amorphous phase.

![Graph showing complex modulus and phase lag](image)

**Figure 4.67** Complex modulus and phase lag of HDPE 7058 (solid lines), HDPE 7108 (dashed lines) and HDPE HM4520XP (dotted lines) measured during three-point bending dynamic mechanical experiments at 1 Hz.

### 4.7 Conclusions

The creep behaviour of HDPE could be described accurately up to strain levels of 5% with the Schapery creep model. Above strain levels of 2%, true stress and true strain need to be
applied. At strain levels higher than 5% yielding starts to occur and this could not be described by the model.

The two-processes model, with two parallel viscoelastic processes with each its own time-stress superposition function, could describe the creep behaviour of HDPE over the entire region, including the yield behaviour.

The Schapery relaxation model could also describe the creep behaviour of HDPE over the entire region.

Predictions of the three models above for monotonous stress histories were reliable for the same time, stress and strain domain as the creep data were fitted well. Generally, the two-processes model gave the best predictions. The complex yielding behaviour of HDPE can only be described accurately by a model with at least two parallel processes.

The three constitutive models proved unreliable in predicting the response of HDPE to non-monotonous stress histories. Although predictions for stress relaxation were good at strain levels below 2%, predictions for recovery failed completely. In case of the Schapery creep model this was due to the absence of viscoplasticity. The two-processes model could not describe the recovery data quantitatively, though qualitative identical recovery behaviour could be obtained by a second fit. The Schapery relaxation model also did not include viscoplasticity. Also the initial recovery stage, which obeyed superposition, was not predicted properly.

With implementation of viscoplasticity into the Schapery creep model the recovery data could be fitted accurately. Predictions for other non-monotonous stress histories also improved by including viscoplasticity.

Creep curves measured at different temperatures showed that HDPE is a thermo-rheologically complex material. However below yielding, i.e., in the region where the Schapery creep model was successful, the creep curves for different temperatures could be superimposed on double logarithmic scale by vertical and horizontal shifting. This means that they can be described by a thermo-rheologically simple model in this region.

The effect of ageing on (iso-age) creep curves could be described by time-ageing time superposition. Additional vertical shifts on logarithmic strain scale seemed necessary. More research is needed for more precise conclusions on ageing of HDPE.
Different grades of HDPE qualitatively showed identical creep behaviour. A larger crystalline fraction results in less creep at short creep times and high stress levels.

4.8 References


5 FEM implementation

5.1 Introduction

The prediction of the mechanical behaviour of a complex plastic product can only be carried out numerically. The Finite Element Method (FEM) is an excellent tool for this and has been applied extensively for numerical analysis of constructions. FEM calculations with viscoplasticity which also require numerical time integration have been applied and reported extensively. Literature on nonlinear viscoelasticity is less extensive and literature on numerical aspects such as stability and computation time is scarce.

For our research the Schapery creep model was extended to account for multiaxial stresses and then implemented into a FEM package. The Schapery creep model could adequately describe the creep behaviour of HDPE up to strain levels of 5%. The FEM package MARC was selected for implementation because of its open structure and its extended capability to handle nonlinear material behaviour.

Different strategies for the numerical integration of the hereditary integrals were tested for their speed and numerical stability during FEM analyses of HDPE products. The validity of the extension of the Schapery creep model to multiaxial stress states was verified by comparison of FEM calculations and experimental results on torsion and three-point bending. Finally the creep response and the response to a constant deformation speed of an HDPE bottle crate were calculated. A fully viscoelastic analysis was compared with a simplified analysis with creep isochrones.

5.2 Schapery creep model for multiaxial stress states

The creep formulation, i.e., strain as a function of the stress, of linear viscoelastic behaviour for multiaxial stress states is given by equations (2.50). These equations imply that the viscoelastic behaviour for each stress/strain direction is independent of the stress and strains of the other directions. Seven directions can be distinguished: one volumetric and six deviatoric.

In case of the nonlinear viscoelastic Schapery creep model, the nonlinearity is accounted for by four Schapery stress functions: $g_0(\sigma)$, $g_1(\sigma)$, $g_s(\sigma)$ and $a_v(\sigma)$, see
section 2.2.2. The Schapery stress functions were assumed to be scalar functions so that substitution of the creep conditions into the Schapery model yields the general expression for multiaxial creep, equation (2.51), see [5.1, 5.2]. This means that the value of, for instance, \( a_e(\sigma) \) holds for all six deviatoric stress directions. In reality this is not necessarily so. Though the assumption results in a large simplification of the three-dimensional model.

The volumetric behaviour was approximated by linear elasticity since for polymers the volumetric response is in general much stiffer than the deviatoric response, see section 2.2.6 and [5.3] for HDPE. Particularly, for thin walled constructions the volumetric contribution to the plane stress behaviour is very small.

![Stress-strain curve](image)

**Figure 5.1** Stress during straining experiments in extension at 23°C with strain rates of approximately 0.01 (○), 0.001 (△) . . . 0.00001 (●) and in compression 0.01 (●), 0.001 (▲) . . . 0.00001 (★). The markers denote the measurements and the (solid/dashed) lines denote the (extension/compression) data transformed into true stress and true strain.
5.2 Schapery creep model for multiaxial stress states

Experimental results of Buckley et al. [5.2] indicated that the nonlinearity during creep depends on the first two invariants $I_1$ and $I_2$, (2.52), of the stress tensor. For the Schapery creep model this means that the Schapery functions $g_0$, $g_1$, $g_2$ and $a_e$ depend on these two invariants. Without loss of generality the second invariant of the stress tensor $I_2$ can be replaced by the second invariant of the deviatoric stress tensor $I_2'$.

The effect of the hydrostatic pressure $p = -\frac{1}{3}I_1$ on the viscoelastic behaviour can be found by comparing extension and compression data. The experiments with a constant cross head speed have already been discussed for the extension case, see section 4.3.1. These data are presented in one graph together with the compression data in figure 5.1. The compression data were obtained by compression of 1 cm prismatic pieces machined from the center of tensile bars between two smooth and flat surfaces, see section 3.3.2. The zero strain reference is inaccurate due to the gradual contact between specimen and moving surface. This was compensated for by shifting the strain data by -0.006.

**Figure 5.2** Stress during straining experiments in compression at 23°C with strain rates of 0.01 (●), 0.001 (△) . . . .0.00001 (*) and the Schapery creep model predictions with measured stress as input (solid lines).
Figure 5.1 shows that the most of the difference between compression and extension data is the result of the effect of true stress and true strain. In extension the true stress is larger than the engineering stress whereas it is smaller for compression. However the lines in figure 5.1 still show a difference between compression and extension. Particularly for the larger strain values these differences can hardly be the result of the inaccuracy of the zero strain level for compression. The higher stiffness for compression, i.e., angle of the lines in figure 5.1, can be caused by the effect of the hydrostatic pressure but also by the increasing true strain rate. In case of compression a constant engineering strain rate causes an increasing true strain rate while in extension the opposite holds. Also the friction between specimen and the smooth surfaces might have played a role.

The Schapery creep model fit was used to predict the compression data. As was observed in figure 4.22 the extension data could be predicted accurately up to strain levels of 6 to 7%. The measured stress during compression as a function of time was taken as input to calculate the strains, since the strain data are less accurate. The results are given in figure 5.2. All but the fastest experiment are again predicted properly up to strain levels of 5 to 6%. Note that the error is determined by the horizontal distance between lines and data. At higher strain levels the Schapery model is, like for extension, inaccurate due to the onset of yielding.

On basis of the adequacy of the predictions of the compression data with the Schapery creep model which was fitted on tensile creep data in section 4.2.1, it was decided to neglect the effect of the hydrostatic pressure on the Schapery stress functions. Implementation of a pressure effect will only lead to a small improvement in case of uniaxial stress states. The apparent absence of a large nonlinear effect of the hydrostatic pressure seems to contradict the many theories on mechanical dilatation induced nonlinearities (rejuvenation/free volume), [5.4].

The Schapery functions were therefore considered to depend on the second invariant of the deviatoric stress tensor only. The model then becomes equivalent to that of Pao et al. [5.5], see equation (2.55):

\[
d_s(t) = \frac{1}{2} J_0 g_0(\tau_{eq})\xi_y + \frac{1}{2} g_i(\tau_{eq}) \int_0^t \Delta J(\psi(t) - \psi(\xi)) \frac{d\left( s_0 g_2(\tau_{eq}) \right)}{d\xi} \, d\xi \\
\psi_m(t) = \frac{1}{2} B_0 \sigma_m
\]

(5.1a)  (5.1b)

with again the reduced times:

\[
\psi(t) = \int_0^t \frac{d\xi}{a_{eq}(\tau_{eq})} \quad \psi(\xi) = \int_0^\xi \frac{d\xi}{a_{eq}(\tau_{eq})}
\]

(5.2)
5.2 Schapery creep model for multiaxial stress states

The equivalent stress was taken equal to the Von Mises stress, see equation (2.53a).

The model above implies that the viscoelastic strain is incompressible and that therefore Poisson’s ratio for the viscoelastic strain equals 0.5. Even though Mears et al. [5.6] measured an increase of the tensile yield stress for PE with the hydrostatic pressure applied, their results imply that the compressive yield stress is only 6 to 7% higher than the tensile yield stress and are therefore not in disagreement with our data. Neglecting any nonlinear pressure effect implies that the model will give bad predictions if relatively large volumetric strains occur.

5.3 FEM implementation of the Schapery creep model

In the previous section the Schapery creep model was extended to account for multiaxial stress situations. In this section the numerical aspects of the FEM implementation are discussed. The user subroutine Hypela offers the MARC user the opportunity to implement nonlinear material behaviour by means of a FORTRAN program. Therefore this subroutine was used for the implementation of the Schapery model.

From earlier research on FEM implementation of viscoplasticity, [5.7, 5.8], it is known that the numerical integration of the constitutive equations, either differential or hereditary integral equations, can be done by forward integration (fully explicit) or by backward integration (fully implicit) or by a combination of both, see section 2.3.1. The first method is easy to program and needs only few computations per time integration step but might suffer from numerical instabilities. Most applications of viscoelasticity and FEM concerned the fully explicit method, [5.3, 5.9 - 5.11]. The second method needs more computations per time step but is numerically stable. Different numerical strategies were programmed to find an optimum for speed and stability. All will be based on the integral approach, see equations (2.62, 2.63, 2.68).

5.3.1 MARC and the subroutine Hypela

The subroutine Hypela enables the user of MARC to write a FORTRAN program for the description of nonlinear material behaviour. The MARC program provides Hypela the strain at the beginning of the step, \( \varepsilon_n \), and the strain increment during the step, \( \Delta \varepsilon \), since like most FEM codes, MARC utilizes the displacement method [5.7, 5.8]. In the material subroutine Hypela the stresses at the end of the step, \( \sigma_{n+1} \), and the material stiffness matrix, \( D_{n+1} \), must be computed and returned to MARC.
Nonlinear elasticity
In figure 5.3 the one-dimensional case for a nonlinear elastic material curve is given. The following equation holds:

$$\sigma_{n+1} = \sigma_n + g + D\Delta \varepsilon$$

(5.3)

with $g$ the stress offset vector.

Figure 5.3  Nonlinear elastic material behaviour.

Since the displacement method is used, the displacement field and hence also the strain increments $\Delta \varepsilon$ are altered by MARC until force equilibrium is obtained, i.e., the unbalanced force vector is within a given margin. For this FEM iteration process, different iteration strategies can be employed [5.7, 5.8]. The material response to changes in the strains is given in the material stiffness matrix. This is visualized as the tangent in the one-dimensional case of figure 5.3. The material stiffness matrices of all integration points of all elements are gathered by MARC and put into the construction stiffness matrix, which together with the unbalanced force vector, determines the displacement field, and hence the strain increments $\Delta \varepsilon$, of the next FEM iteration step. The material stiffness matrix may deviate from the exact material stiffness matrix as long as convergence of the FEM iteration process is not lost. After convergence the next step is started.

Nonlinear viscoelasticity
In case of nonlinear viscoelastic behaviour figure 5.4 is obtained. The effect of a time integration step is shown. Even if $\Delta \varepsilon = 0$ the stress offset $g$ is nonzero due to stress relaxation. Hence $g$ will from now on be called the stress relaxation vector. In case of a
constant stress, \( \sigma_{n+1} = \sigma_n \), the strain increment \( \Delta \varepsilon \) can be called the creep vector. If \( t_{n+1} \) is increased a curve with a larger stress relaxation vector and a larger creep vector is obtained. The curvature is the result of the nonlinearity in the viscoelastic behaviour.

MARC also has an automatic time stepping option which chooses the time integration steps \( \Delta t_n = t_{n+1} - t_n \) such that stress and strain increments are below a given maximum. This means however that during the FEM iteration process, the material curve of figure 5.4 also changes and consequently the number of FEM iterations necessary for force equilibrium can increase. Therefore it was decided to prescribe the time steps for all calculations beforehand and to choose time steps which are sufficiently small to assure that the numerical time integration of the stress history is accurate enough.

One important requirement of our FORTRAN program for the subroutine Hypel is that it can be applied to the most important elements of the MARC element library. In our case the plane stress and shell elements are most important since many plastic constructions are thin walled. All these elements have in common that stresses in certain directions are zero and drop out. The strains in these directions are not zero due to lateral contraction but are unimportant to the problem and can therefore be neglected (except when large lateral contractions lead to an important increase of the true stress).

![Diagram](image)

**Figure 5.4** Nonlinear viscoelastic material behaviour.

The omission of certain stresses hardly results in any difficulties for our creep model since it is stress based. The corresponding strains are just not evaluated, which saves memory and computation time. However in case of a relaxation model the corresponding strains are needed to evaluate the equivalent strain which determines the Schapery or other nonlinearity functions. In case of a relaxation model one is forced to use the “tensile modulus-Poisson’s ratio approach” which was also used by Lai et al. [5.9] although it was
applied to a creep model. The relaxation form of the "bulk modulus-shear modulus" approach, see equations (5.1), will inevitably lead to large programming difficulties if it is to be applicable for plane stress and shell elements as well.

5.3.2 Fully explicit time integration

For this method, a forward integration step ($\alpha = 0$) is combined with forward values for the elasticity Schapery parameters, $g_o$ and $g_1$, see equations (2.62, 2.63, 2.68). The result is that the stresses and strains depend on each other linearly, see figure 5.5. The material stiffness matrix is the elastic stiffness matrix multiplied by $g_o$. The stress relaxation vector $g$ and the stiffness matrix $D$ are independent of the strain increment $\Delta \epsilon$ and therefore have to be calculated only once, resulting in a relatively few computations. However numerical instabilities will arise when the time step size exceeds a critical value [5.7, 5.12, 5.13], or when $\Delta \sigma$ is large.

![Figure 5.5](image-url)  

**Figure 5.5** Fully explicit time integration step on a nonlinear viscoelastic material.

5.3.3 Implicit time integration

The time integration step now also contains a backward part which makes it nonlinearly dependent on the end stress values. The most straightforward method would be as in equation (2.68): the solution is the sum of $(1-\alpha)$ times the forward solution and $\alpha$ times the backward solution. However there is a more efficient way:
\[ Q_{k,ij}^n = q_{k,ij}^n + \left( J_k x_{ij}^n g_{ij}^n - q_{k,ij}^n \right) \left( 1 - \exp \left( -\frac{(1-\alpha) \Delta t_n}{\tau_k a_{eq}^n} \right) \right) \quad (5.4a) \]

\[ q_{k,ij}^{n+1} = Q_{k,ij}^n + \left( J_k x_{ij}^{n+1} g_{ij}^{n+1} - Q_{k,ij}^n \right) \left( 1 - \exp \left( -\frac{\alpha \Delta t_n}{\tau_k a_{eq}^{n+1}} \right) \right) \quad (5.4b) \]

A forward step with time step size \((1-\alpha)\Delta t_n\) is followed by a backward step with time step size \(\alpha\Delta t_n\), see also figure 5.6 for visualization. The major advantage is that the backward step and the forward step of the next time integration step can be combined and evaluated as one step, since the incremental formulation of the integral is exact for stepwise loading histories, see [5.14] and section 2.3.1. The next expression therefore holds:

\[ Q_{k,ij}^{n+1} = Q_{k,ij}^n + \left( J_k x_{ij}^{n+1} g_{ij}^{n+1} - Q_{k,ij}^n \right) \left( 1 - \exp \left( -\frac{\alpha \Delta t_n + (1-\alpha) \Delta t_{n+1}}{\tau_k a_{eq}^{n+1}} \right) \right) \quad (5.5) \]

The strains \(d_{ij}^{n+1}\) at time \(t_{n+1}\) are expressed in terms of \(Q_{k,ij}^{n+1}\), which represent the convolution integral of the Schapery creep model, see equations (2.62, 2.63). After a successful integration step (force equilibrium achieved) the integrals are updated in terms \(Q_{k,ij}^{n+1}\), so that the backward time step \(\alpha \Delta t_n\) and the forward time step \((1-\alpha)\Delta t_{n+1}\) are combined as one step. Still \(\alpha = 0\) yields forward integration and \(\alpha = 1\) backward integration.

**Figure 5.6** Combined forward backward integration step.

For a nonlinear viscoelastic material, figure 5.7 is obtained for the stress strain relation. With fixed time step size the end strains that the creep model calculates solely depend on the stresses at the end, since the stresses at the beginning of the time step are
fixed. In other words the tangent approximation of the nonlinear stress-strain relation is a function of $\Delta \varepsilon$, see figure 5.7. However the displacement method for FEM calculations provides the material subroutine with the end strains. With nonlinear behaviour an iteration process within Hypela is needed to find the end stresses for which the given end strains are found by the creep model. Therefore a damped quasi-Newton iteration with Broyden’s method [5.15, 5.16] for updating the inverse of the Jacobian was programmed in the subroutine Hypela to invert the creep model.

In case of plasticity the implicit method becomes unconditionally stable if $\alpha \geq \frac{1}{2}$ [5.7, 5.17]. In our case this transition to stability might not be situated exactly at $\frac{1}{2}$. The value of $\alpha$ should be chosen as small as possible without occurrence of instabilities since a lower value of $\alpha$ results in more explicit integration and hence in a more linear stress strain relation. The more linear this relation the less FEM and Hypela iterations are needed.

![Diagram showing implicit time integration step on a nonlinear viscoelastic material.](image)

**Figure 5.7** *Implicit time integration step on a nonlinear viscoelastic material.*

In addition to the stresses at the end of the step $\sigma_{n+1}$ the FEM program also needs the material stiffness matrix $D$, which can be seen as the tangent to the stress strain curve at the end stress. But since we have a creep model only the compliance matrix $C$ can be evaluated:

$$\Delta \varepsilon = C \Delta \sigma$$  \hspace{1cm} (5.6)

This matrix needs to be inverted to obtain the stiffness matrix:

$$\Delta \sigma = C^{-1} \Delta \varepsilon = D \Delta \varepsilon$$  \hspace{1cm} (5.7)
Not only the inversion process is time consuming but also the evaluation of all the derivatives with respect to all stress directions has to be done for the compliance matrix.

Furthermore this nonlinear model will as a rule result in a non-symmetric material stiffness matrix. For instance, in case of a tensile stress a stress increment in the tensile direction yields a Von Mises stress increment, which differs from the Von Mises stress increment in case of a stress increment in an other direction. This then also holds for the material nonlinearities which depend on the Von Mises stress.

However a good alternative approximation for the material stiffness matrix is available which needs no numerical matrix inversion and no calculations of all derivatives. If it is assumed that all Schapery stress functions do not change during the time integration step then this step becomes linearly viscoelastic and superposition is valid. One can also state that the nonlinear Schapery parameters are taken in a forward manner and therefore fixed. The approximate material stiffness matrix becomes:

\[
D = \begin{pmatrix}
K_0 + \frac{4}{3}G_a & K_0 - \frac{2}{3}G_a & K_0 - \frac{2}{3}G_a & 0 & 0 & 0 \\
K_0 - \frac{2}{3}G_a & K_0 + \frac{4}{3}G_a & K_0 - \frac{2}{3}G_a & 0 & 0 & 0 \\
K_0 - \frac{2}{3}G_a & K_0 - \frac{2}{3}G_a & K_0 + \frac{4}{3}G_a & 0 & 0 & 0 \\
0 & 0 & 0 & 2G_a & 0 & 0 \\
0 & 0 & 0 & 0 & 2G_a & 0 \\
0 & 0 & 0 & 0 & 0 & 2G_a
\end{pmatrix}
\]  

(5.8)

with:

\[
K_0 = \sqrt{B_0}, \quad G_a = \sqrt{J_a}
\]

(5.9)

and:

\[
J_a = D_0k_0 + g_1g_2 \sum J_k \left( 1 - \exp \left( \frac{-\alpha \Delta t_k}{\tau_a a_{eq}} \right) \right) + g_1g_2 \frac{\alpha \Delta t_k}{\eta a_{eq}}
\]

(5.10)

The material stiffness matrix provided to the FEM program as well as the inverse Jacobian for the start of the quasi-Newton iteration within Hypela are approximated as shown above. This expression for the stiffness matrix \( D \) is far more simple than those derived in [5.3, 5.9, 5.10]. Furthermore no numerical matrix inversion is needed, saving a large amount of computation time.
5.3.4 Semi-explicit time integration

The time integration, see equations (5.4) and (5.5), for this method is done with all the Schapery parameters taken in a forward manner. The stepping parameter $\alpha$ still determines whether the (linear) stress is determined forwards or backwards or by combination of both. The result is that model response is linear viscoelastic each time integration step. Hence the stress-strain behaviour is linear again and the stress relaxation vector $g$ and the material stiffness matrix $D$ are independent of the strain increment $\Delta \epsilon$, see figure 5.8. A similar approach for the numerical solution of a nonlinear viscoelastic model with differential equations was used by Dooling et al. [5.18].

The material stiffness matrix is the same as the approximate stiffness matrix for the implicit method, see equations (5.8, 5.10). Since the material stiffness matrix contains a viscoelastic part it depends on the time step size. As a result the two straight lines of figure 5.8 are not parallel (as in figure 5.5), but since it was decided to prescribe the time step sizes beforehand $g$ and $D$ are constant for each the time step integration. Therefore no iteration process within the material subroutine Hypela is needed.

This method is very similar to the fully explicit method and hence will be computationally inexpensive. However, this method will give an exact implicit step when a linear viscoelastic material is dealt with. Consequently numerical instabilities are not caused by time step size, as is the case for the fully explicit method, but by the change of the nonlinear parameters that would appear if a fully implicit time step were taken. Since at low stress levels the viscoelastic behaviour is linear, numerical instabilities might occur at high stress levels only.

![Figure 5.8](image)

**Figure 5.8** Semi-explicit time integration step on a nonlinear viscoelastic material.
5.3.5 Semi-implicit time integration

The larger calculation times of the fully implicit method are caused by the iterations needed within the material subroutine Hypela and by the larger number of FEM iteration steps needed to achieve force equilibrium. This larger number of FEM iterations is the result of the nonlinearity of the material subroutine response, which is not present in case of an explicit integration scheme.

Figure 5.9  *Semi-implicit time integration step on a nonlinear viscoelastic material with FEM iteration 1 and 2.*

An accurate iteration loop within Hypela can be regarded as a waste of computation time when the FEM iteration process is in the beginning and far off the end solution. If it is assumed that during each FEM iteration step the nonlinear Schapery parameters are constant then no iterations are needed within Hypela since the response is linear viscoelastic again, see figure 5.9. By recalculation of the nonlinear Schapery parameters at the start of each FEM iteration an iteration process is obtained which after convergence will yield the implicit solution, since the Schapery parameters are also converging to those of the implicit solution. In fact the two iterations are reduced to only one. This will lead to a reduction of computation time if the number of FEM iterations does not increase. One has to bear in mind that one FEM iteration step is much more computationally expensive than one iteration step within Hypela.

Material updating strategies in FEM analyses with nonlinear material behaviour are common practice in case of plasticity, [5.7, 5.8].
5.3.6 Calculations using creep isochrones

Many engineering design problems where viscoelastic behaviour of plastics plays a role are creep related. Most designers then perform FEM calculations using creep isochrones [5.19]: the response of a product to a constant load at time $t$ is evaluated using the stress-strain relation found by the creep curves at that time $t$. See figure 4.2 for the creep isochrones of HDPE. The FEM analysis is then similar to a nonlinear elastic analysis. Sometimes even this nonlinear stress-strain relation is linearized [5.19, 5.20].

In case of linear viscoelastic behaviour stepwise loading of a complex product yields a stress distribution which does not alter in time. Consequently, every part of the product has a stepwise stress history. This was first acknowledged by Alfrey [5.21]. Calculations using creep isochrones are exact when no stress distributions in time are present. Krishnaswamy et al. [5.22] showed that this is true for nonlinear viscoelastic creep behaviour of the separable form:

$$\varepsilon(t) = D(t)G(\sigma)$$  \hspace{1cm} (5.11)

Note that our Schapery creep model fit of the creep data, see equation (4.1) is only for a linear strain identical to equation (5.11). Stress redistributions are probably small and the calculations with creep isochrones are then a good approximation for the true creep response.

The implementation of creep isochronous response in the material subroutine Hypela is very straightforward. When no stress redistributions are present a backward implicit variant of equations (5.4, 5.5) gives the exact material response for all time step values. Calculations with creep isochrones are performed simply by setting $Q_{i,j} = 0$. The effect of the stress history has then disappeared and it is no longer necessary to calculate many time integration steps to obtain the creep response at large creep times. The time step size can be set equal to the creep time. Note that calculations with creep isochrones will be very complicated and uneconomical when a relaxation model is applied.

Calculations with creep isochrones were already performed for tensile straining and stress relaxation. The results were given in figures 4.24 and 4.35.

5.4 FEM predictions for experiments

All FEM calculations were performed with the Schapery creep model fit explained in section 4.2.1. All 11 Kelvin-Voigt elements needed to fit the creep data were used. Since the fits were made with the constant volume assumption the elastic bulk compliance was set to 0.000045 1/MPa, leading to an elastic Poisson’s ratio of 0.488.
The finite element analyses were performed with the displacement method [5.7, 5.8] using the Full Newton-Raphson procedure for convergence to force equilibrium. The accuracy for this iteration was set to 1% relative error of the residual forces. The MARC option for large displacements was used to account for the large geometrical nonlinearities. Also the option for configuration updating, resulting in true stress and true strain for the subroutine Hypela, was used.

The time step sizes were always prescribed in the input file and not determined by MARC. For the creep calculations a time decade was split into 16 steps which are equidistant on logarithmic time scale. Calculations were done in double precision. Storage of the $Q_{k,ij}$, stresses, strains and stiffness parameters were done in single precision to save memory. Calculations were performed on a pentium I 200 MHz with 256 MB SD-RAM.

In case of the semi-explicit method the stepping parameter $\alpha$ was set equal to 1 so that the linear part of the "excitation" is taken backward: $\sigma g_1 (\sigma) = \sigma_{\epsilon1} g_1^\epsilon$, because this will result in maximum stability. The equivalent stress and the Leaderman (Schapery integral) function $g_2 (\sigma)$ were updated after the evaluation of all $Q_{k,ij}$. The semi-implicit and the implicit method were also used in combination with backward integration.

### 5.4.1 Routine check calculations

A number of FEM calculations were carried out with only one element. Figure 5.10 shows the measured creep strain of HDPE as markers and the strain calculated with the 8-node solid element type (el. nr. 7). The semi-explicit method was applied here. As can be observed all FEM results are very close to the measurements for strains below 5%, just as the Schapery creep model fit itself, see figure 4.4.

The 8 MPa creep curve was also determined with the fully explicit, implicit and the semi-implicit method. No deviations from the measurements were found. However, the fully explicit calculation broke off at a creep time of 36517 s and with a time increment of 5652 s, see section 5.6.

Finally different element types were applied with the semi-explicit method: plane stress, thick shell, thin shell and truss element (el. nr. 3, 75, 139 and 9). The true cross section needed for the evaluation of the true stress was done by MARC with the constant volume assumption. Furthermore the Herrmann (incompressible) solid element was used (el. nr. 84). This element yields faster calculations for constant volume analysis. For all analyses the calculated creep curves coincide with the curves of figure 5.10.
Figure 5.10  Creep strain of HDPE during creep experiments at load levels of 1 (v), 2 (v), 4 (o), 6 (●) . . . . 18 (♦) MPa and the FEM results of one 8-node brick element using the Schapery creep model (lines).

5.4.2 Three-point bending creep of a beam

Small tensile bars were machined from injection moulded square plates and tested for their tensile creep response at 23°C and 32 weeks after injection moulding, see section 3.3.6. The bars were cut either parallel or perpendicular to the injection moulding direction. The results from tensile creep experiments with these bars are shown in figures 5.11 and 5.12 as markers. Later the Schapery creep model fits to these data were used to predict the creep response during three-point bending of prismatic beams. These beams were microtomed from the square plates such that their width is equal to their thickness, i.e., thickness of the plates.

The experimental data were fitted up to 5% strain with the Schapery creep model. The Leaderman function $g_2(\sigma)$ of the fit of section 4.2.1 was used. The spectrum was set equal to the spectrum of section 4.2.1 multiplied by an (initially) unknown factor. This unknown factor and the elastic compliance were the only two parameters which were varied in order to fit the creep data. The Schapery fit of the creep curves are also displayed in figures 5.11 and 5.12. The “parallel” and “perpendicular” fit, shown together in figure 5.12, are situated so close together that anisotropy seems negligible.
Figure 5.11 Strain of HDPE during tensile creep experiments on bars machined from plates parallel to the injection moulding direction at load levels of 2 (▲), 4 (○), 6 (●) ... 18 (♦) MPa and the Schapery creep model fit (lines).

Figure 5.12 Strain of HDPE during tensile creep experiments on bars machined from plates perpendicular to the injection moulding direction at load levels of 2 (▲), 4 (○), 6 (●) ... 18 (♦) MPa and the Schapery creep model fit (dashed lines) and as comparison the fit of the "parallel" data (solid lines).
**Figure 5.13**  *FEM mesh for the three-point bending calculations ($\frac{1}{4}$ beam); grey scale denotes the vertical displacement.*

Prismatic beams, also machined from the square plates, were subjected to a three-point bending deformation with a constant force in the center of 5.886 N (600 grams) while the center displacement was monitored in time. See section 3.3.6 and figure 3.6 for the experimental setup. In case the engineering bending theory were valid, this load would yield a maximum stress of 9.5 MPa.

FEM predictions for the three-point bending response were performed using the material parameters found by the Schapery fits shown in figures 5.11 and 5.12. Only one quarter of the beam was modelled because of symmetry, see figure 5.13 for the mesh. Half of the length of the beam was divided into 17 elements and half of the width was divided into 2 elements. The 4-node thick shell elements (el. nr. 75) which were used had 9 layers in the thickness direction. The forces were applied directly upon the corresponding nodes, i.e., no contact option was used. The backward semi-explicit method was applied.

The experimental results of three-point bending experiments are shown together with the FEM predictions in figure 5.14. The open markers show the experimental results from in-plane bending. In case of in-plane bending the skin and the core have the same strain distribution. As can be observed the FEM results are in good agreement with these data. The accuracy of the predictions confirms the assumption that the effect of the hydrostatic stress is minimal. Calculations were also performed with 11 layers instead of 9
layers and with 8-node thick shell elements (el. nr. 22) instead of 4-node elements. In both cases differences with the shown FEM results were less than a few percent.

![Graph showing center displacement vs creep time](image)

**Figure 5.14** Displacement of the center during three-point bending creep of HDPE beams cut parallel (△, ▽) and perpendicular (●, ○) to the injection moulding direction together with the FEM predictions with the Schapery creep model for "parallel" (solid line) and "perpendicular" (dashed line); open markers denote in-plane bending and solid markers out-of-plane bending.

The data resulting from out-of-plane bending exhibit a larger creep strain and hence a lower stiffness. In case of out-of-plane bending the skin is in the outer fiber with the highest strains and the core is around the neutral axis with the lowest strains. It can therefore be concluded that the skin has a lower stiffness than the core. The material behaviour obtained from tensile experiments can therefore give a too stiff prediction for plate bending.
5.4.3 Torsional creep of a tube

Hollow cylinders were injection moulded and tested for their tensile and torsion creep response at 23°C and 32 weeks after production, see section 3.3.6. The tensile creep data were fitted with the Schapery model and this model was used for FEM predictions of the torsion creep response. Material attributed deviations can therefore only be the result of the skin core effect, see previous section.

The procedure for fitting the creep curves in section 5.4.2 was repeated. The spectrum for the Schapery fit was set equal to that found during the fitting of the injection moulded tensile bars, multiplied by an (initially) unknown factor, see section 4.2.1. The Leaderman function was also set equal to the one found for the tensile bars. The unknown factor and the elastic compliance were used to fit the creep data up to strain levels of 5%. The experimental results together with the fit are shown in figure 5.15. It can be observed that the description of creep is satisfactory.

The FEM calculations for the response of the HDPE tube to a constant torque moment were done with 4-node axisymmetric elements with twist (el. nr. 20). Only one half of the tube needed to be modelled because of antisymmetry. Also the part of the tube which was clamped was included in the FEM calculations, so that possible shear creep inside the clamps is included. The metal parts which clamped the specimen were considered infinitely stiff during the FEM analysis. Figure 5.16 shows the mesh. As before the backward semi-explicit method was applied with 16 time integration steps per decade. The MARC option for constant volume analyses was not used since then convergence to force equilibrium could not be achieved after two time steps. These two steps, however, yielded identical results as without the option.

The angle of the complete tube as a function of the creep time is given in figure 5.17 for the measurements as well as for the FEM simulation. The deviations between experiment and simulation are rather large, especially for small time values. This can partly be explained by the value of Poisson’s ratio. The relation between elastic shear and tensile stiffness is as follows:

\[ G_0 = \frac{E_0}{2(1+\nu_0)} \]  \hspace{1cm} (5.12)

An elastic Poisson’s ratio lower than 0.5 will yield a higher elastic shear stiffness and hence result in better agreement between torsion measurements and predictions. However the deviations at creep times of 10 seconds cannot be fully attributed to this effect.
Figure 5.15 Strain of HDPE during tensile creep experiments on tubes at load levels of 1 (▼), 2 (▲), 4 (○), 6 (●) . . . . 16 (○) MPa and the Schapery creep model fit (lines).

Figure 5.16 FEM mesh for the torsion calculations (1/2 cylinder); grey scale denotes the shear stress (dark area clamped).
If the nonlinearity of the viscoelastic model, i.e., the Leaderman function $g_2(\sigma)$, does not solely depend on the equivalent stress but also on the hydrostatic stress then the response to torsion or shearing will be stiffer due to the absence of a hydrostatic stress. Consequently the torsion angle will be smaller and closer to the data. However the effect will be small at creep times around 10 seconds since this would not affect the (linear) elastic response. Moreover the difference in the slope of the creep curves of the data and the model in figure 5.17 will only increase by this. Therefore deviations cannot be explained simply by a nonlinear effect of the hydrostatic stress. Also attribution of the nonlinearity to the maximum shear stress instead of the equivalent shear stress does not improve predictions for torsion.

![Graph showing angle vs. creep time](image)

Figure 5.17  Angle of HDPE during torsion creep experiments on tubes at load levels of 0.39 (.), 0.79 (•), 1.2 (○) and 1.5 (●) Nm and the FEM predictions (lines).

In figure 5.18 the creep angle rate during the creep experiments and their FEM predictions are shown. It can be observed that the creep angle of the fourth curve (●) is overestimated while the creep angle rate is underestimated. Therefore presence of slip at the clamping of the specimens or other experimental errors are unlikely to be responsible for the deviations between experiment and prediction. The presence of a softer skin as was found during three-point bending experiments, see the previous section, will only result in more
torsion creep than expected from tensile data and can consequently not be held responsible for the deviations.

The FEM calculations were also performed with 8-node Herrmann elements (el. nr. 66) and with a finer mesh, but both results were close to the FEM results shown in figure 5.17. Loading of the tube in more steps with a smaller load increment did not change the FEM predictions either.

![Graph](image)

**Figure 5.18** Angle rate of HDPE during torsion creep experiments on tubes at load levels of 0.39 (○), 0.79 (•), 1.2 (○) and 1.5 (●) Nm and the FEM predictions (lines).

It seems that the extension of a one-dimensional nonlinear viscoelastic model to a three-dimensional model is not so straightforward as expected. In case of three-point bending the shear deformation was negligible with respect to the bending deformation ($l/h > 5$) and hence only the tensile and compressive stresses needed to be taken into account. Extension of the one-dimensional model to three dimensions was therefore not strictly necessary and problems related to this extension to multiaxial stress states were absent.
5.4.4 Compression of a dish

An injection moulded axisymmetric dish with 150 mm diameter and 3 mm average thickness, see figure 3.5 for the radial cross section, was compressed in the center with different constant displacement speeds, see section 3.3.6 for the experimental set up. FEM predictions of the dish response to compression were made using the material parameters which were found by fitting the creep response of the injection moulded tensile bars with the Schapery creep model, see section 4.2.1. Therefore, differences in the injection moulding process between the bars and the dishes were not accounted for.

![FEM mesh for the dish calculations (1/2 dish); grey scale denotes the compressive displacement.](image)

Axisymmetric 2-node thick shell elements (el. nr. 1) with 9 layers per element were used to model the dish, see figure 5.19 for the mesh. Since three-point bending results showed that the difference between calculations with 9 or 11 layers is less than one percent, the 9 layers were considered sufficient.

The displacements of the outer node were restrained since the dish was put on a flat metal base with a ring encircling the brim of the dish. The radial displacement of the center node was restrained to avoid problems due to negative radii. The distance between the center node and the next node of the dish was 2.375 mm and the imposed compressive
displacement was applied to this next node. It is assumed that this boundary condition is a sufficiently accurate simulation of the experiment where the dish was compressed in the center by a 12 mm diameter cylinder rounded off with a 40 mm radius. Furthermore the stresses in the center of the dish are so high that the Schapery creep model is no longer valid, see section 4.2.1.

![Graph showing force vs. displacement](image)

**Figure 5.20** Reaction force of the HDPE dish during compression experiments at rates of 1 (▲), 0.1 (●), 0.01 (○) and 0.001 (●) mm/s and the FEM predictions with the semi-explicit method (irregular lines) and the fully implicit method (smooth lines).

The FEM model of the dish was compressed 14 mm with 8 time integration steps per mm. The calculations were first performed with the fully explicit method. This method did not give satisfactory results since calculations broke off, see also section 5.6. Even for compression at the highest rate and time increments of 0.125 s, the calculations stopped after 50 increments at a 6.25 mm compressive displacement.

The measurements and the FEM simulation results with the backward semi-explicit method are shown in figure 5.20. As can be observed the data are well predicted up to 7 mm compression. At larger displacements deviations occur probably due to the simplification of the force application. Small numerical instabilities first appeared at 9 mm compression,
where nonlinearities due to the high stresses at the center of the dish are starting to dominate. However calculations continued and therefore the stability of this integration scheme was in our case considered to be satisfactory.

To overcome the stability problems the backward semi-implicit method was also applied. The results are not shown in figure 5.20 since the method failed to converge to force equilibrium when the dish was compressed more than approximately 2.3 mm, see also section 5.6. Finally the backward implicit method was used. As shown in figure 5.20 the numerical stability problems completely disappeared.

Analyses of the stresses in the dish showed that during compression not only uniaxial tension and compression stresses due to bending occurred, but also large circumferential stresses appeared. Stress/strain situations were comparable to plane strain situations. FEM calculations with 3-node elements (cl. nr. 89) deviated only a few percent from those with 2-node elements.

![Figure 5.21](image)

**Figure 5.21**  *FEM mesh for the bottle crate calculations (1/4 crate); grey scale denotes the stress in vertical (z) direction.*
5.5 FEM analyses with a bottle crate

An HDPE bottle crate was investigated for its creep response and its response to compression with a constant displacement speed. The material parameters from the Schapery creep model found during the fit of the creep data on injection moulded tensile bars, see section 4.2.1, were applied in the FEM calculations.

Only one quarter of the crate needed to be modelled, because of the presence of two symmetry planes. Antisymmetric deformation modes were therefore not considered. The mesh, delivered by MARC, is given in figure 5.21. The mesh consisted of 1523 4-node thick shell elements (el. nr. 75) with 9 layers. To simulate a stack of bottle crates the vertical displacement of the entire top edge was set to zero while a uniform vertical displacement on the bottom edge was imposed. In case of creep the total force on the bottom was constant. In case of compression the displacement of the bottom was increased with a constant speed in time.

![Graph showing creep time vs. displacement](image)

**Figure 5.22** Compressive displacement of \( \frac{1}{4} \) bottle crate during creep with average stress levels of (-) 1, 2, 3, 4 and 5 MPa (up in graph) calculated by the FEM model with full viscoelastic analyses (solid lines) and analyses with creep isochrones (dashed lines).
5.5.1 Full viscoelastic analyses

The bottle crate was loaded with a constant total force simulating creep due to a dead weight load. The FEM results are presented in figure 5.22 as solid lines. The crate was also compressed 4 mm in 80 equal time steps at different constant speeds. The solid lines in figure 5.23 show the results of these calculations. Both calculations were done with the backward semi-explicit method. No instabilities occurred and calculation times were acceptable, see also section 5.6. An average stress of 1 MPa along the edge of the quarter bottle crate corresponds to a total force of 525 N.

During the creep analysis time-delayed buckling of the crate occurred when the average stress level exceeded 1 MPa. This buckling is seen as an upward swing in the deformation load curve of figure 5.22. A higher stress results in a shorter time before buckling. In case of a constant compression speed, buckling of the crate always occurred before at less than 4 mm compressive displacement. The compression speed only affects the total force needed for the compression.

![Graph showing force vs. displacement](image)

**Figure 5.23** Compressive total force of 1/4 bottle crate during compression at constant rates of 0.5, 0.05, 0.005 . . . 0.0000005 mm/s (down in graph) calculated by the FEM model with full viscoelastic analyses (solid lines) and analyses with creep isochrones (dashed lines).
5.5.2 Viscoelastic analyses using creep isochrones

The accuracy of calculations on creep of products with creep isochrones was verified by comparing them with full viscoelastic calculations. The calculations with the bottle crate were therefore repeated but this time with the material behaviour found by the creep isochrones, see figure 4.2. The implicit backward integration scheme was used, since the Schapery parameters must be taken backwards.

The results of the analyses are shown in figures 5.22 and 5.23 as broken lines. The differences in case of creep of the bottle crate are rather small indicating that stress redistributions are minimal. The response with creep isochrones is somewhat less stiff from the onset of buckling. For the prediction of the buckling behaviour of a product this means that by performing calculations using creep isochrones a conservative estimate is made. Results of the compression analyses show that differences between creep isochrones and full viscoelastic calculations are no longer negligible, see figure 5.23, although errors remain below 15%. The results are in agreement with those of figure 4.24.

5.6 Numerical stability and computation time

Fully explicit method

From the results on creep calculations with one single element it can be concluded that the fully explicit method is unreliable concerning the numerical stability, see section 5.4.1. The 8 MPa creep calculations broke off at a time step of 5652 seconds and a creep time of 36517 seconds due to failure of the Gaussian elimination process in MARC. This is probably caused by the high (elastic) material stiffness matrix $$\mathbf{D}$$ and the large amount of creep during the time step which resulted in an extremely large stress relaxation vector $$\mathbf{g}$$, see figure 5.5. The time step size is, however, still large with respect to the retardation time of the fastest Kelvin-Voigt element, i.e., 0.01 seconds, which is a time measure for the response rate of the nonlinear viscoelastic strain to a stress increment [5.23].

Calculations with compression of a dish with the same material parameters and numerical strategy broke off at small time increments, see section 5.4.4, again due to failure in the Gaussian elimination process. For compression at the highest rate, for instance, where the time step size was 0.125 seconds, calculations stopped after 50 increments at a compressive displacement of 6.25 mm. The apparent stability of the fully explicit method during creep and the unreliability during calculations with varying stresses can be explained as follows. A forward (explicit) integration step is exact and equals a backward (implicit) step if the stress is constant, which is the case during creep, but not during compression.
 Implicit method
The implicit method was always applied with a stepping parameter $\alpha = 1$, resulting in backward (fully implicit) calculations. No numerical instabilities were ever encountered with this method. The effect of a lower stepping parameter, for instance $\alpha = \frac{1}{2}$, was not investigated. Calculation times were however larger than with the semi-explicit method, see also the next item.

 Semi-explicit method
This method was always applied with $\alpha = 1$ to obtain maximum stability, since then the linear part of the “excitation” function is taken backwards: $\sigma_{e2}(\sigma) = \sigma_{e1}(\sigma)^{n}$. Calculations with three-point bending of a beam, torsion of a cylinder, compression of a dish and creep and compression of a bottle crate, showed that this method is very reliable though not infallible. Results with the dish showed that numerical instabilities can occur when large material nonlinearities are present, though they were minimal for our calculations, see section 5.4.4.

In general instabilities are less during creep calculations, since for constant stresses the forward and backward step are identical for a creep model. The compression of the dish was performed with a constant displacement speed resulting in almost constant strain rates throughout the dish. Since the first strain increment $\Delta \varepsilon$, that MARC provides to Hypela during iteration for force equilibrium, is based on the assumption of constant strain rate, an improvement of the stability with minimal extra computations can be achieved. If for each first call to Hypela the time integration step is solved implicitly and after that the nonlinear Schapery parameters are assumed constant, as in the semi-explicit case, then stability for constant strain rate cases will improve. The only additional computational effort will be one iteration process within Hypela for every time integration step. This possibility has not been tested yet.

The calculation times of the backward semi-explicit method were compared with those of the backward implicit method. The latter method was applied using creep isochrones. Calculation times will hardly be affected by this, since the only difference from full viscoelastic analyses was the updating of the internal parameters $Q_{k,0}$. The results for creep are shown in figure 5.24 and the ones for compression in figure 5.25.

As can be observed in figure 5.24 the average computation time increases with the creep load while the calculations carried out within the material subroutine Hypela are independent of the load level for the semi-explicit method. Therefore this increase was caused by the larger amount of FEM iterations needed to find force equilibrium. This is mainly caused by buckling which started earlier at higher load levels and therefore gives a relatively larger contribution to the total computation time. The material nonlinearity during
the creep analysis with an average stress of 1 MPa played such a minor role that the semi-explicit and the implicit method gave equal computation times.

![Graph showing average creep stress vs. computation time](image)

**Figure 5.24** Total computation time divided by the number of time integration steps of the FEM calculations on compressive creep of \( \frac{1}{4} \) HDPE bottle crate using the backward semi-explicit method (light grey) and the fully implicit method (creep isochrones, dark grey).

In case of compression the computation time per increment hardly depends on the compression speed since buckling occurs irrespective of the compression speed. Figure 5.25 shows that implicit calculations took approximately 20% more computation time than with the semi-explicit method. This is caused partly by the iteration process within the material subroutine Hypela and partly by the larger amount of FEM iterations for force equilibrium. The latter is caused by the nonlinearity in the material response with respect to the strain increment \( \Delta \varepsilon \), compare figures 5.7 and 5.8. The computation times for compression, however show no dependence on the stress level, i.e., compression rate. Apparently the effect of larger material nonlinearities at higher stresses and the effect of smaller time steps at higher stresses resulting in less nonlinear creep per increment, compensated each other.

One also has to bear in mind that during our calculations the Full Newton-Raphson iteration process [5.7, 5.8] was chosen for MARC to obtain force equilibrium. This means that during each iteration the construction stiffness matrix is recalculated. This was necessary for the buckling analyses of the bottle crate. In case no buckling or other phenomena with nonlinearities caused by large displacements or large strains occurs, the
semi-explicit method will be even more computationally advantageous than the implicit method. The semi-explicit method will then yield a simple linear problem for every time integration step whereas the implicit method then still yields a nonlinear problem. However, since the approximation of the material stiffness matrix, see equations (5.5 - 5.7), was successful during calculations with the fully implicit method, both methods can be used in combination with the Modified Newton-Raphson iteration process, [5.7, 5.8], which is faster in this case.

![Graph showing average comp. time per incr. (s) vs. compression rate (mm/s)](image)

**Figure 5.25** Total computation time divided by the number of time integration steps of the FEM calculations on compression of ¼ HDPE bottle crate using the backward semi-explicit method (light grey) and the fully implicit method (creep isochrones, dark grey).

**Semi-implicit method**

The semi-implicit method, also applied with $\alpha = 1$, was not successful. Calculations with the dish stopped already after few time steps due to divergence of the force equilibrium error during FEM iteration. Apparently the merging of two iteration processes into one does not result in a numerically stable single iteration process. Even if the method would work the computation time reduction with respect to the implicit method would be less than obtained by the semi-explicit method, i.e., 20%.
5.7 Conclusions

The Schapery creep model was extended to account for multiaxial stress states by assuming that all Schapery functions depend on the Von Mises stress only. The volumetric response was assumed linear elastic. Comparison of tension and compression data of HDPE showed that the nonlinear effect of the hydrostatic stress is small and therefore negligible.

Injection moulded (square) plates of HDPE bent out-of-plane exhibited 7 to 15% more creep than when bent in-plane. Apparently the skin of the plate is softer than the core. Predictions based on the material behaviour from tensile experiments can therefore be unreliable for predictions of plate bending.

Four numerical strategies for the numerical time integration of the Schapery model were implemented into the FEM package MARC: fully explicit method, implicit method, semi-explicit method and semi-implicit method.

The fully explicit method and the semi-implicit method turned out to be unreliable regarding the numerical stability.

The implicit method with backward integration (fully implicit) did not suffer from any numerical instabilities.

The semi-explicit method is based on the assumption that the Schapery function values are constant during the time integration step. Since the material model response becomes linear viscoelastic during each time integration step, no iterations within the material program are needed. Also the material stiffness matrix can be calculated directly without matrix inversion. The semi-explicit method was in our case approximately 20% faster than the backward implicit method. Small numerical instabilities occurred only when material nonlinearities were large.

The material stiffness matrix as obtained from the semi-explicit method can be applied successfully for the implicit method as an approximation of the true stiffness matrix.

FEM analyses with creep isochrones as material behaviour turned out to be a good alternative for full viscoelastic calculations when the creep response of HDPE products is to be obtained.
5.8 References


6 Viscoelasticity and designers

6.1 Introduction

The importance of weight reduction of products is increasing because of the growing economical and environmental pressure. Consumer products are often designed with plastics and, therefore, thorough knowledge of the time dependent mechanical behaviour of plastics is required for minimal weight design. HDPE is a polymer, which is also used extensively in load bearing applications such as bottle crates and containers.

This thesis is primarily concerned with the modelling of the nonlinear viscoelastic behaviour of HDPE and the numerical implementation of this behaviour for FEM analyses. Many results from the constitutive modelling can be applied to other polymers. The numerical aspects of the FEM implementation of the Schapery creep model are of such a general nature that they are applicable for any nonlinear time dependent material behaviour.

HDPE was chosen for the constitutive modelling because of its pronounced creep behaviour and its applications in load bearing products like containers and bottle crates. Three different models were used to describe the creep behaviour of HDPE. In this research the validity and the ease of handling of the different models have been determined. The choice between a simple engineering model with separation of variables and more complex models depends on the specific requirements of the designer.

The numerical work on FEM calculations has resulted in different time integration strategies. Their numerical stability and computational efforts have been investigated. For the determination of a product with a time dependent stiffness by means of FEM calculations, the designer can choose between the fast and simple semi-explicit method and the more complicated but unconditionally stable (fully) implicit method. Furthermore, the validity of simplified FEM analyses using creep isochrones, which lead to nonlinear elastic analyses, has been examined.

6.2 Experimental results on HDPE

The creep measurements on injection moulded tensile bars at 23°C covered a total time span of 8 weeks. The effect of ageing during these measurements was not significant. The creep
curves exhibit characteristic patterns, which have also been found for other polymers like LDPE and PP, see section 4.1. The creep curves of HDPE show a pronounced convex curvature at creep times between 1000 and 10000 seconds. This curvature appears at the same creep time for all stress levels below 10 MPa (strains below 5%). Stress relaxation data also exhibit a (convex) curvature at relaxation times of approximately 1000 s. The creep strain depends on the creep stress in a nonlinear way. At short creep times this nonlinearity is smaller.

At strain levels above 5% the pattern of the creep curves changes. The strain rate during creep decreases with time (primary creep), but, in case of stresses above 10 MPa, is followed by a stabilization (secondary creep) and eventually an increase of the strain rate (tertiary creep). This is due to yielding. The yield points obtained by creep and tensile straining experiments exhibit complex behaviour when plotted in an Eyring plot, see figure 4.10. Also creep curves obtained at different temperatures cannot always be superimposed on a master curve, indicating that HDPE is thermo-rheologically complex. At least two different viscoelastic processes are responsible for the constitutive behaviour of HDPE.

For comments on the effect of grade and injection moulding conditions on the viscoelastic behaviour see section 3.6: creep of different grades of HDPE.

6.3 Constitutive modelling

First of all two categories of viscoelastic models can be distinguished: creep models (strain as a function of stress) and relaxation models (stress as a function of strain). In case of linear viscoelasticity the models are equivalent and can be considered merely as a change of input and output. The choice of model then depends solely on the type of phenomena to be described: the creep model is most suitable to describe creep and the relaxation model is most suitable to describe relaxation.

In this research the material parameters of a model were determined by fitting of the creep data. Three different constitutive models were investigated: the Schapery creep model, the two-processes model and the Schapery relaxation model. The Schapery creep model is an engineering model which, has been applied widely to describe nonlinear viscoelasticity. The Schapery relaxation model is in its formula equivalent to the Schapery creep model but with stress and strain interchanged. The two-processes model is a more complicated model, which consists of two nonlinear viscoelastic processes in parallel. Each process is identical to the Leonov model, which is a subset of the Schapery creep model. The model is able to describe rheologically complex viscoelastic behaviour. The experimental creep data show that HDPE is rheologically complex.
Schapery creep model

The creep strain of HDPE could be described by the sum of a linear elastic strain and a nonlinear viscoelastic strain, which is the product of a time function and a nonlinear stress function, i.e., separation of variables. See section 4.2.1. Therefore only one of the four Schapery functions were needed. Expressing the model in terms of true stress/strain instead of engineering stress/strain extended the validity of the model from strain levels of 2% to 5%. At strain levels above 5% (complex) yielding occurred and this cannot be described by one single Schapery creep process.

Predictions with the Schapery creep model for monotonous stress histories (tensile straining and ramp loading) are in agreement with data as long as time, stress and strain are in the same region as where the creep data could be fitted accurately.

Predictions for non-monotonous stress histories (recovery, ramp unloading after loading and stress relaxation) are not in agreement with data due to the presence of viscoplasticity. The initial recovery obeys superposition and is predicted properly, but with progressing recovery time a plastic strain emerges, which is not accounted for by the Schapery creep model. After implementation of viscoplasticity the recovery data could be fitted properly, see section 4.3.2. The obtained model gave good predictions for ramp unloading. Stress relaxation was already predicted properly up to strain levels of 2%. At higher strain levels, however, the stiffness was overestimated. Yielding during stress relaxation at strain levels of 8 and 16% is the main reason for this. The situations where the Schapery creep models are valid, are given in figures 6.1 and 6.2.

**Figure 6.1** Validity of the Schapery creep model for HDPE.

**Figure 6.2** Validity of the Schapery creep model with viscoplasticity for HDPE.
Two-processes model
This model assumes that the viscoelastic behaviour of HDPE is determined by two parallel nonlinear viscoelastic processes making it rheologically complex. Each process is assumed to obey time-stress superposition (horizontal shifting of creep curves). The peculiarity of this model is that it predicts vertical shifts of the creep curves on logarithmic strain scale in the region of no yielding. The model also yields a convex curvature in the creep curves and complex (double) yield behaviour, see figures 4.9 and 4.10. This is all in agreement with our measurements, see section 4.2.2.

Two fits were made of the creep data. For fit 1 the two time-stress superposition functions were "free" whereas for fit 2 (section 4.3.2) they were restricted to the Eyring form. With fit 1 the creep data were accurately described over the entire region. With fit 2 the data could not be fitted correctly. Because the model consists of two processes, more numerical computations were needed to calculate the response to stress or strain histories. Also fitting of creep data becomes very complicated.

Predictions with the two-processes model (fit 1) for monotonous stress histories are in agreement with data. The complex yield behaviour measured during the tensile straining experiments is also predicted sufficiently accurate.

In case of non-monotonous stress histories the two-processes model, like the Schapery creep model, fails. In case of stress relaxation the predictions are good up to 2% strain but at higher strain levels the stiffness is overestimated, though less than for the Schapery creep model due to the proper prediction of yielding. The ramp unloading data are also predicted properly but the recovery data are completely in error. The validity of the two-processes model (for our fit) is given in figure 6.3. Fit 2 was made to prove that the recovery behaviour could, at least qualitatively, be described by the two-processes model.

![Diagram](image)

**Figure 6.3**  *Validity of the two-processes model for HDPE.*
6.3 Constitutive modelling

Schapery relaxation model
This model is based on the observation that the nonlinear effect in stress relaxation can be accounted for by a vertical shift of the relaxation curve on logarithmic stress scale, see section 4.2.3. This model was used to fit the creep data, but since this model is a relaxation model more numerical computations were needed to calculate its creep response than for the Schapery creep model. Although this model cannot describe Eyring type of yielding behaviour, the yielding could be described reasonably well by the strain softening term in this model. The creep data below 10% strain were fitted properly.

Predictions with the Schapery relaxation model for monotonous stress histories are in reasonable agreement with data. The yield behaviour found by the tensile straining experiments is not predicted accurately.

Predictions for non-monotonous stress histories are not in agreement with data due to absence of viscoplasticity in the model. The model also predicts faster initial recovery than expected from superposition. This is also in disagreement with the data. Only stress relaxation is predicted properly when strain levels are below 2%. The regions where the model is valid, are shown in figure 6.4.

![Figure 6.4](image_url)  
**Figure 6.4**  
*Validity of the Schapery relaxation model for HDPE.*

The effect of temperature on creep
Creep data at various temperatures ranging from 18 to 43°C proved that HDPE is a thermo-rheologically complex material, see figure 4.51. However, the inability to superimpose creep curves at various temperatures occurs only in the transition region from no yielding to yielding. Therefore below strain levels of 5%, the region most relevant for designers, the effect of temperature on creep can be modelled with a thermo-rheologically simple model, i.e., shifting of a master curve. The creep data below 5% strain could indeed be fitted
properly with the Schapery creep model with time-temperature and strain-temperature superposition. The shifting values could be described by the Arrhenius equation.

The effect of ageing on creep
The injection moulded tensile bars showed relatively little ageing; relatively 10 to 15% less strain when the ageing time increases from 2 weeks to 128 weeks, see section 4.5. Hence the effect of ageing for creep stresses of 2 and 4 MPa was in the order of experimental error. Analysis of the data seems to indicate that creep curves at various ages can be superimposed by vertical and horizontal (time-ageing time superposition) shifting on double logarithmic scale. Since the maximum creep time for ageing experiments was only two hours, no conclusions can be drawn for creep with much longer creep times.

Creep of different grades of HDPE
Creep measurements were performed on three different grades of HDPE, see section 4.6. Two injection moulding grades with different molecular weights and one extrusion grade were examined. All three grades exhibit the same qualitative creep behaviour. This implies that, though grade and processing conditions can alter the quantitative material response, the same models can be applied to different grades of HDPE.

Since in our case all injection moulding conditions were similar and chosen such to minimize anisotropy and internal stresses, the effects of these conditions seemed small. However, in practice injection moulding cycle times are much smaller and larger variations in creep can be encountered.

The difference in creep behaviour between the two injection moulding grades was negligible. The extrusion grade exhibited a stiffer behaviour at high stress levels and short creep times. Results from DMTA measurements showed that the extrusion grade of HDPE had a higher melt temperature than the injection moulding grades, which indicates that it had a more stable crystalline phase.

6.4 FEM implementation
The Schapery creep model was chosen for implementation into the FEM code MARC, since this model has been used extensively for the modelling of creep or creep/recovery. The model can account for many nonlinear effects and includes the Leaderman creep model and the Leonov creep model as subsets. When used in combination with a spring dashpot model for the creep compliance function it allows fast numerical calculations by means of recurrent relations. Storage of the strains in all individual dashpots is necessary and in fact allows the "forgetting" of the preceding stress/strain history and the use of fast numerical integration
with recurrent expressions, see section 2.3.

The Schapery model was extended to account for multiaxial stress states by assuming that the shear deformation is nonlinear viscoelastic and the volumetric deformation linear elastic. Because of the small difference between tension and compression curves of HDPE, obtained by applying a constant strain rate, the nonlinearity was assumed to depend on the Von Mises stress only. Also HDPE was assumed to be incompressible. For most plastic products these simplifications in case of plane stress situations, i.e., thin walled constructions.

The FORTRAN program for the implementation of the Schapery creep model into MARC can handle many different FEM element types, such as solid, plane stress, thin shell and thick shell elements. This is necessary since most plastic products are thin walled, which causes certain stresses to be absent.

Four numerical time integration strategies were programmed into the material subroutine Hypela: fully explicit method (widely applied), implicit method, semi-explicit method and semi-implicit method, see section 5.3. Furthermore the program offers the opportunity to perform calculations using creep isochrones. The calculation of the creep response of a plastic product is often approximated by designers by means of an analyses with creep isochrones. The analysis is then simplified to a nonlinear elastic analysis.

FEM predictions for experiments
For the verification of the extension of the one-dimensional Schapery model to the three-dimensional form, experimental results with non-uniaxial stress states were compared with FEM predictions. The experiments performed were: three-point bending creep of a beam, torsion creep of a hollow tube and compression of a dish.

Predictions of the three-point bending data were accurate. Experiments with three-point bending showed that the skin and the core of injection moulded plates have different stiffnesses; the core is stiffer. Creep results of tensile bars machined parallel and perpendicular to the injection moulding direction, showed a small degree of anisotropy. Predictions of the torsion data were poor. Only part of the deviations could be explained by compressibility of the (assumed incompressible) viscoelastic strain. Compression data of the dish, in which the strain situation is comparable to the plane strain situation, were predicted properly.

Numerical stability and computation time
From the FEM calculations it can be concluded that the fully explicit method suffers from numerical instability. Although a fully explicit time step requires fewest computations per time step, the numerical instability forces the use of many small time steps, which in overall makes this method computationally inefficient. The (fully) implicit method is completely
numerical stable but requires more calculation time due to an iteration process within the material subroutine Hypela. The backward semi-explicit method is very stable. Only in case of large nonlinearities, instabilities can occur. The computation time for one time step is (almost) equal to that of a fully explicit step, but the increased stability allows larger time steps. Therefore it can be concluded that this is a very efficient method. The semi-implicit method failed due to non-convergence.

**Analyses using creep isochrones**

In analyses with creep isochrones the material stress-strain relation at a specific loading time is obtained by the creep curves at that same time. A complete evaluation of the stress history is then no longer necessary. FEM creep analyses with an HDPE bottle crate showed that deviations between full viscoelastic calculations and calculations using creep isochrones are negligible. Therefore, stress redistributions during creep were for this case minimal. In case of compression with a constant displacement speed the stress is no longer constant and deviations emerge. In our case the stress of the full viscoelastic analyses is underestimated by 15% or less. The stress relaxation curves obtained from full viscoelastic analyses with the Schapery creep model (not FEM), however, differ only a few percent from those obtained with creep isochrones. Simplification using creep isochrones is therefore also allowed for stress relaxation problems (with yielding absent).

### 6.5 Conclusions

In case of monotonous stress histories the constitutive behaviour of HDPE can be modelled with the engineering Schapery creep model as long as strains remain below 5%. Above 5% strain (complex) yielding commences and the material stiffness decreases fast. In case of non-monotonous stress histories (unloading) viscoplasticity needs to be accounted for.

The two-processes model can accurately describe creep of HDPE including the complex yielding behaviour. However an accurate quantitative description of the material behaviour could not be obtained in case of non-monotonous stress histories. Its complexity and the large amount of computations needed with this model makes it at the moment unsuitable for engineering purposes.

The Schapery relaxation model is an engineering model, but more difficult to use for fitting of creep data.
In the region where the Schapery creep model is valid, i.e., strains below 5%, the effect of temperature on creep can be described by shifting of a master curve, i.e., a thermo-rheologically simple model.

FEM calculations on large products can be performed efficiently with semi-explicit time integration. In case of large material nonlinearities, numerical instabilities can occur. These can be avoided by performing fully implicit time integration.

In case of creep and in case of stress relaxation (no yielding) full viscoelastic FEM analyses can be replaced by analyses with creep isochrones. This simplifies the analysis to a nonlinear elastic analysis.
Schapery creep model parameters

Sub Modelspectrum()
    Dim i As Integer
    M = 11
    ReDim TAU(M), D(M)
    For i = 1 To M
        TAU(i) = 10 ^ (i - 3)
    Next i
    D0 = 0.000629
    D(1) = 0.0000103
    D(2) = 0.0000401
    D(3) = 0.0000605
    D(4) = 0.000128
    D(5) = 0.000257
    D(6) = 0.000389
    D(7) = 0.000361
    D(8) = 0.000201
    D(9) = 0.000283
    D(10) = 0.000298
    D(11) = 0.000352
End Sub

Function G2func(s)
    Dim sabs
    sabs = Abs(s)
    G2func = (1 + 0.237 * sabs + 0.000136 * sabs ^ 2 + 0.000026 * sabs ^ 3 + 0.0000181 * sabs ^ 4) * s
End Function

Function G2barfunc(s)
    Dim sabs
    sabs = Abs(s)
    G2barfunc = (0.9001 + 0.219 * sabs) * s
End Function

Function G3barfunc(s)
    Dim sabs
    sabs = Abs(s)
    G3barfunc = (0.0999 + 0.018 * sabs + 0.000136 * sabs ^ 2 + 0.000026 * sabs ^ 3 + 0.0000181 * sabs ^ 4) * s
End Function
Two processes model parameters (fit 1)

Sub ModelSpectra()
    Dim i As Integer
    NA = 21
    NB = 9
    ReDim TAU(A(NA), DA(NA))
    ReDim TAU(B(NB), DB(NB))
    For i = 1 To NA
        TAU(i) = 10 ^ (i - 3)
    Next i
    For i = 1 To NB
        TAU(i) = 10 ^ (i - 3)
    Next i
    DB = 0.001207
    DB = 0.003194
    DA(1) = 0.0000631
    DA(2) = 0.00007586
    DA(3) = 0.0000912
    DA(4) = 0.0001155
    DA(5) = 0.0001495
    DA(6) = 0.0002008
    DA(7) = 0.0002644
    DA(8) = 0.000333
    DA(9) = 0.0004001
    DA(10) = 0.0004648
    DA(11) = 0.0005333
    DA(12) = 0.0006199
    DA(13) = 0.0007472
    DA(14) = 0.0009518
    DA(15) = 0.001295
    DA(16) = 0.001886
    DA(17) = 0.002916
    DA(18) = 0.0047
    DA(19) = 0.007691
    DA(20) = 0.01236
    DA(21) = 0.01882
    DB(1) = 0.000005985
    DB(2) = 0.00003796
    DB(3) = 0.0001906
    DB(4) = 0.0007581
    DB(5) = 0.002387
    DB(6) = 0.005953
    DB(7) = 0.01176
    DB(8) = 0.01838
    DB(9) = 0.02277
    ETA = 1E+25
    ETAB = 217500
End Sub
Function asignaa(s)
    Dim a2, a3, a4, a5, a6, a7, a8, a9, a10, sabs, dummy
    sabs = Abs(s)
    If sabs > 15 Then
        asignaa = 1 / Exp(41.0775 + 15.025 * (sabs - 15))
    Else
        a2 = -10.53260112
        a3 = 15.14993295
        a4 = -7.865389576
        a5 = 2.191232778
        a6 = -0.360027002
        a7 = 0.035897273
        a8 = -0.002129919
        a9 = 0.0000690083
        a10 = -0.00000937385
        dummy = 1 / Exp(a2 * sabs + a3 * sabs^2 + a4 * sabs^3 + a5 * sabs^4 + a6 * sabs^5 + a7 * sabs^6 + a8 * sabs^7 + a9 * sabs^8 + a10 * sabs^9)
        If dummy > 1 Then
            asignaa = 1
        Else
            asignaa = dummy
        End If
    End If
End Function

Function asignab(s)
    Dim a2, a3, a4, a5, a6, a7, a8, sabs, dummy
    sabs = Abs(s)
    If sabs > 12 Then
        asignab = 1 / Exp(4.18639 + 0.57262 * (sabs - 12))
    Else
        a2 = -0.436154547
        a3 = 0.668175778
        a4 = -0.242756021
        a5 = 0.045947308
        a6 = -0.004740251
        a7 = 0.000251617
        a8 = -0.0000535494
        dummy = 1 / Exp(a2 * sabs + a3 * sabs^2 + a4 * sabs^3 + a5 * sabs^4 + a6 * sabs^5 + a7 * sabs^6 + a8 * sabs^7)
        If dummy > 1 Then
            asignab = 1
        Else
            asignab = dummy
        End If
    End If
End Function
Schapery relaxation model parameters

Sub Model Spectrum()
    Dim i As Integer
    M = 12
    ReDim TAU(M), E(M)
    For i = 1 To M
        TAU(i) = 10^((i - 3))
    Next i
    E0 = 11.6
    E(1) = 0
    E(2) = 329
    E(3) = 225
    E(4) = 376
    E(5) = 223
    E(6) = 168
    E(7) = 94.3
    E(8) = 52
    E(9) = 32
    E(10) = 20
    E(11) = 15.8
    E(12) = 362
End Sub

Function H2func(e)
    Dim eabs, a2, a3, a4, a5, a6, a7, a8, a9, a10
    eabs = Abs(e)
    a2 = 48.17616682
    a3 = -528.1949647
    a4 = 9068.333151
    a5 = -142302.415
    a6 = 2148544.832
    a7 = -21617028.98
    a8 = 123209351.9
    a9 = -362232764.7
    a10 = 428559571.2
    If eabs < 0.2 Then
        H2func = e / (1 + a2 * eabs + a3 * eabs^2 + a4 * eabs^3 + a5 * eabs^4 +
                      a6 * eabs^5 + a7 * eabs^6 + a8 * eabs^7 + a9 * eabs^8 + a10 * eabs^9)
    Else
        H2func = 0.026312138 + (e - 0.2) * 0.02
    End If
End Function
Two processes model parameters (fit 2)

Sub Modelsapectra()
    Dim i As Integer
    NA = 59
    NB = 11
    ReDim TAU(A(NA), DA(NA))
    ReDim TAUB(NB), DB(NB)
    For i = 1 To NA
        TAU(i) = 10 ^ (i - 3)
    Next i
    For i = 1 To NB
        TAUB(i) = 10 ^ (i - 3)
    Next i
    DOA = 0.001873888
    DOB = 0.000821005
    DA(1) = 0.0000350749
    DA(2) = 0.0000363688
    DA(3) = 0.000377103
    DA(4) = 0.000391014
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End Sub

Function Afunca(s)
    Dim sabs
    sabs = Abs(s)
    If (sabs / 0.098371) < 0.0001 Then
        Afunca = 1
    Else
        Afunca = (Sinh(sabs / 0.098371)) / (sabs / 0.098371)
    End If
End Function
Function Afuncb(s)
    Dim sabs
    sabs = Abs(s)
    If (sabs / 1.9814) < 0.0001 Then
        Afuncb = 1
    Else
        Afuncb = (Sinh(sabs / 1.9814)) / (sabs / 1.9814)
    End If
End Function
Summary

Nonlinear Viscoelastic Behaviour of HDPE
Constitutive Modelling and Finite Element Method Implementation

The mechanical response of plastic products is time dependent because of the viscoelastic nature of the material. A plastic chair loaded with a person will not deform only elastically, but it will also exhibit creep: a viscoelastic deformation, which will slowly increase in time. The nonlinear viscoelastic behaviour of plastics cannot be ignored for minimal weight design of load carrying plastic products.

The subject of this research is to model the nonlinear time dependent material behaviour of HDPE, a thermoplastic used for instance in bottle crates, containers, pipe lines and packaging. The material description had to be implemented into a FEM package so that the time dependent mechanical behaviour of complex shaped plastic products can be evaluated numerically.

The viscoelastic behaviour of HDPE is determined by many factors: stress level, stress history, temperature (history), time after production and the HDPE grade. Therefore it was decided to reduce the number of experiments by excluding combined effects. This means that the effect of ageing (time after production) is investigated at one temperature only.

The tensile creep behaviour of an injection moulding grade of HDPE at 23°C with negligible ageing during creep was investigated first. Creep experiments with a maximum creep time of eight weeks were performed at various stress levels. The creep strain was measured up to strain levels of 20%.

The creep data were described accurately up to 5% strain by the Schapery creep model. Above 2% strain the use of true stress and true strain is recommended. The Schapery creep model could not describe the (complex) yielding behaviour, which occurred at strain levels above 5%.

The two-processes model, which is aimed to describe rheologically complex viscoelastic behaviour, could model the creep data over the entire strain region. Also the yielding behaviour was described properly.

The Schapery relaxation model could model the creep data over the entire strain region as well, though not as accurately as with the two-processes model. Particularly
yielding was described less accurately, since a relaxation model cannot describe Eyring type of yielding.

In case of monotonous (increasing) stress histories all three models gave good predictions in the strain region where the creep data were fitted well. The Schapery creep model failed above strain levels of 5 to 6% whereas the Schapery relaxation model could describe the behaviour reasonably accurate above these strain levels. Only the two-processes model could predict yielding above these strain levels accurately.

In case of non-monotonous (increasing and decreasing) stress histories all three models gave bad predictions. In case of recovery after creep both the two-processes model and the Schapery relaxation model failed. The initial recovery however was predicted properly by the Schapery creep model, but the subsequently emerging plastic strain was not included. The stress during stress relaxation at strain levels of 0.5, 1 and 2% was predicted accurately. At higher strain levels all models overestimated the stress.

The effect of viscoplasticity was included into the Schapery creep model and the recovery data were used for fitting. Predictions for other non-monotonous stress situations improved.

The two-processes model was used to fit the creep data a second time but with different parameters. It was shown that this second fit describes the measured recovery data qualitatively, including the occurrence of a viscoplastic strain. Unfortunately a quantitative fit of the creep and recovery data could not be obtained.

Creep measurements at temperatures of 18, 23, 28 . . . . 43°C showed that HDPE is thermo-rheologically complex. This means that the effect of temperature cannot be described over the entire region by shifting of a master creep curve on double logarithmic scale. However below 5% strain, the region where the Schapery creep model is valid, the creep curves could be superimposed on a master curve.

The effect of ageing on creep of our HDPE was small and at small stress levels the effect was in the order of the experimental error. However the data indicated that the creep curves could be superimposed by shifting on double logarithmic scale.

Creep data on three different HDPE grades showed qualitatively identical creep behaviour. In case of two injection moulding grades with different average molecular masses but with equal crystallinities, the data are even quantitatively the same. The extrusion grade with the higher crystallinity exhibited less creep at high stress levels.

The Schapery creep model was chosen for implementation into the FEM package MARC, since its has been used widely to describe nonlinear viscoelasticity and other models, like the Schapery creep model, are a subset of this model. An optimal numerical treatment of the Schapery creep model requires the description of the creep compliance function to be done with spring dashpot analogy.
The one-dimensional Schapery creep model was extended to account for multiaxial stress states by assuming that the bulk behaviour is linear elastic and decoupled from the deviatoric behaviour. The nonlinearity in the deviatoric components was assumed to depend on the Von Mises stress only.

Four different numerical strategies were programmed: fully explicit, implicit, semi-explicit and semi-implicit method. In case of the fully explicit method forward time integration was performed. This classical method however led to numerical instabilities when large time steps were applied. The implicit method was used with backward time integration (fully implicit) and did not suffer from any instability. However, because of the iteration process needed within the material subroutine more computations were needed per time integration step than for the fully explicit method. With the semi-explicit method, it was assumed that the Schapery functions, which cause the nonlinearity, are constant during the time integration step. Consequently a time step becomes linear viscoelastic and no iterations within the material subroutine are needed. The number of computations needed per time step is comparable to that needed for the fully explicit method. Numerical instabilities due to large time steps were absent for the backward semi-explicit method. The semi-implicit method yielded a non-converging system and was therefore unsuccessful.

Predictions of the Schapery creep model for experiments with stress situations other than uniaxial tension were in agreement with the data in case of bending but not in case of torsion. Only part of the deviations for torsion could be explained by the effect of compressibility. The bending data revealed the presence of a skin and core with different stiffnesses, the skin layer being less stiff.

FEM calculations with a bottle crate with full viscoelastic analyses were compared with simplified analyses using creep isochrones. In case of creep deviations were negligible, indicating that stress redistributions were minimal. In case of stress relaxation at small strain levels (no effect of plasticity), deviations were in the order of a few percent, even though the stress is not constant. In case of compression with a constant displacement speed the simplification is no longer allowed.

Johan G.J. Beijer
Samenvatting

Niet-lineair Visco-elastisch Gedrag van HDPE
Materiaalgedragmodellering en Eindige-Elementen-Methode Implementatie

De mechanische respons van de meeste plastic producten is tijdsafhankelijk vanwege het visco-elastische karakter van het materiaal. Een plastic stoel belast met het gewicht van een persoon, vertoont niet alleen een elastische vervorming, maar zal ook kruipe vertonen: een in de tijd toenemende visco-elastische vervorming bij constant belasting. Het niet-lineaire visco-elastische gedrag van kunststoffen kan niet buiten beschouwing worden gelaten indien men een belast product op minimaal materiaalgebruik wil ontwerpen.

Het doel van dit onderzoek was het modelleren van het niet-lineaire tijdsafhankelijke materiaalgedrag van HDPE, een thermoplast dat onder andere gebruikt wordt in kratten, containers, pijpleidingen en verpakkingen. De materiaalbeschrijving dient vervolgens in een eindige elementen pakket te worden geïmplementeerd zodat de mechanische respons van producten met complexe vormen numeriek bepaald kan worden.

Het visco-elastische gedrag van HDPE wordt bepaald door vele factoren waaronder de belastingsniveau, belastinggeschiedenis, temperatuur (geschiedenis), ouderdom van het materiaal en het type HDPE. Er is besloten om alleen een enkel effect per keer te beschouwen. Zo is bijvoorbeeld het effect van veroudering op kruipe alleen bij één temperatuur bekeken.

Het kruipegedrag in trek van gespuitgiette HDPE trekstaven bij 23°C en een ouderdom van 32 weken werd als eerste onder de loep genomen. Daartoe zijn acht weken durende kruipproeven uitgevoerd met verscheidene spanningsniveaus. De kruipeprek werd tot rekniveaus van maximaal 20% gemeten.

De meetresultaten van de kruipe metingen konden nauwkeurig worden beschreven tot rekniveaus van 5% met het Schapery kruipe model. Boven rekken van 2% levert gebruik van ware rek en ware spanning een betere beschrijving. Het vloeige drag dat optrad boven rekniveaus van 5% kon niet met dit model worden beschreven.

Het twee-processen-model, bedoeld voor het modelleren van rheologisch complex materiaalgedrag, kon de kruipegevens nauwkeurig beschrijven voor alle gemeten rekniveaus. Ook het (complex) vloeige drag kon met dit model gemodellerd worden.
Het Schapery relaxatiemodel was ook in staat de kruipdata over het hele rekgebied te beschrijven, echter niet zo nauwkeurig als het twee-processen-model. Vooral het vloeigedrag werd minder nauwkeurig beschreven, omdat een relaxatiemodel geen Eyring vloeigedrag beschrijven kan.

In geval van een monotone (toenemende) spanningsverloop gaven alle drie de modellen goede voorspellingen in het rekgebied waar de kruipdata ook goed beschreven zijn. Het Schapery kruiemodel faalde boven rekniveaus van 5 à 6% terwijl het Schapery relaxatie model dan nog een redelijke beschrijving opleverde. Alleen het twee-processen-model kon het vloeigedrag bij trekproeven goed voorspellen.

In het geval van een niet-monotone (toe- en afnemende) spanningsverloop gaven alle drie de modellen slechte voorspellingen. Bij kruip gevolgd door “recovery” faalden zowel het twee-processen-model als het Schapery relaxatiemodel. De beginfase van “recovery” werd wel goed voorspeld door het Schapery kruiemodel, maar de plastische rek die vervolgens optrad kon niet worden voorspeld. De spanning tijdens spanningsrelaxatie bij rekniveaus van 0,5, 1 en 2% rek werd wel goed voorspeld door alle modellen. Bij grotere rekken lieten alle modellen een overschatting van de spanning zien.

Het effect van visco-plasticiteit werd vervolgens in het Schapery kruiemodel geïmplementeerd, waarbij de “recovery-data” gebruikt werden voor het bepalen van de modellparameters (“fitten”). Voorspellingen voor niet-monotone spanningsgeschiedenissen verbeterden hierdoor.

Het twee-processen-model was een tweede keer gebruikt om de kruipegevens te beschrijven, echter nu met andere parameters. Voorspellingen voor “recovery” waren met deze tweede “fit” wel kwalitatief in overeenstemming met de metingen, inclusief de viscoplastische rek. Echter een goede beschrijving van zowel kruip en “recovery” kon met het twee-processen-model niet worden verkregen.

Kruipmetingen bij temperaturen van 18, 23, 28 . . . 43°C toonden aan dat HDPE thermo-rheologisch complex gedrag vertoon. Dit houdt in dat het effect van temperatuur op kruip niet beschreven kan worden voor alle rekniveaus met het “schuiven” van een “master-curve”. Echter beneden 5% rek, de regio waar het Schapery kruiemodel geldig was, konden de kruipkrommes wel gesuperponeerd worden op een “master-curve”.

Het effect van “ageing” op kruip van onze HDPE was klein en bij lage spanningsniveaus was het effect in de orde van de experimentele fout. De meetresultaten lijken aan te duiden dat kruipkrommes met verschillende ouderdommen gesuperponeerd kunnen worden op dubbellogaritmische schaal.

Kruipdata van drie types van HDPE vertoonden kwalitatief hetzelfde kruipgedrag. In geval van de twee spuitgiet types met verschillende gemiddelde moleculargewichten, maar met gelijke kristalliniteit, waren de metingen zelfs kwantitatief hetzelfde. Het extrusie type met een hogere kristalliniteit vertoonde minder kruip bij hoge spanningen.
Het Schapery kruipmodel werd verkozen voor implementatie in het eindige-elementenpakket MARC, omdat dit model veel gebruikt is bij het beschrijven van niet-lineaire viscoelasticiteit en omdat het andere modellen, zoals het Schapery kruipmodel, omvat. Een efficiënte numerieke aanpak van het Schapery kruipmodel vereist het gebruik van veerdemper modellen voor het beschrijven van de kruipcompliantiefunctie.

Het een-dimensionale Schapery kruipmodel was uitgebreid voor drie-dimensionale spanningssituaties door aan te nemen dat de volumetrische respons lineair elastisch is en ontkoppelt van de deviatorische respons. Er werd aangenomen dat de niet lineariteit in de deviatorische componenten alleen van de Von Mises spanning afhankt.

Vier verschillende numerieke strategieën werden geprogrammeerd in MARC: volledig expliciete methode, impliciete methode, semi-expliciete methode en de semi-impliciete methode. De volledig expliciete methode gaat gepaard met voorwaardse integratie. Deze klassieke methode resulteerde echter in numerieke instabiliteiten bij te grote tijdsstappen. De impliciete methode werd gebruikt in combinatie met achterwaardse integratie (volledig impliciet) en ging niet gebukt onder enige numerieke instabiliteit. Echter vanwege het nodige iteratieproces binnen het materiaalprogramma zijn meer berekeningen nodig per tijdsstap dan met de volledig expliciete methode. Bij de semi-expliciete methode wordt aangenomen dat de Schapery functies, welke de niet-lineariteit van het model bepalen, constant zijn gedurende de tijdsstap. Als gevolg is het gedrag tijdens de tijdsstap lineair visco-elastisch en zijn er geen iteraties binnen de materiaalsubroutine meer nodig. Het aantal berekeningen nodig per tijdsstap is vergelijkbaar met het aantal van de volledig expliciete methode. Numerieke instabiliteiten ten gevolge van grote tijdsstappen zijn bij de terugwaardse semi-expliciete methode verdwenen. De semi-impliciete methode resulteerde in een niet-convergerend system en was daarom niet succesvol.

Voorspellingen van het Schapery kruipmodel voor experimenten met spanningssituaties anders dan eenassige trek zijn in overeenstemming met de data in geval van buiging, maar in geval van torsie is er een verschil. Het effect van compressibiliteit kan dit verschil slechts gedeeltelijk verklaren. De data over buiging laten zien dat er een verschil in stijheid is tussen huid- en kermateriaal, waarbij de huid minder stijf is.

Eindige-elementen-berekeningen aan een krat met volledige berekening van viscoelasticiteit werden vergeleken met versimpelde berekeningen uitgaande van kruipisochronen. In geval van kruip waren de afwijkingen tussen beide methodes verwoordbaar, hetgeen impliceert dat herverdeling van de spanning minimaal was. Verschillen tussen beide methodes zijn bij spanningsrelaxatie ook kleiner dan een paar procent. Bij compressie met een constante verplaatsingssnelheid was de vereenvoudiging niet meer geldig.

Johan G.J. Beijer
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Curriculum Vitae

Johan Beijer was born on 13 August 1971 in the vivid hart of the Netherlands: Tiel. After finishing high school (VWO) in 1989 in Breda, he studied Aerospace Engineering at the Delft University of Technology. He specialized in the field of materials and constructions. In 1994 he graduated on the subject of buckling of anisotropic plates with mixed boundary conditions under supervision of Professor Vogelesang.

In 1995 he started a Ph.D. research as an AIO (Assistent In Opleiding) at the Delft University on the subject of nonlinear viscoelastic behaviour of polymers, fulfilling his desire for mathematical modelling. During his Ph.D. several educational tasks were accomplished and the course Top Opleiding Polymeer en of the Foundation Polymer Technology Netherlands (PTN) was followed successfully.
List of publications


