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Modelling of creep behaviour in injection-moulded HDPE

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Abstract

Tensile creep experiments on HDPE with a duration of 8 weeks were performed at a temperature of 23°C. Since the tests were performed 32 weeks after moulding of the specimens, the effect of ageing during creep can be neglected. The creep strain can be fitted up to 5% using the Leaderman model in terms of true stress and true strain. However, the yielding at larger strain levels cannot be described with this model. A good fit of the creep data, including yield behaviour, is obtained with a model of two processes in parallel, each having its own time–stress shifting function. Predictions of this model for constant strain rate experiments show good agreement with observations.

Keywords: HDPE; Creep; Leaderman model

1. Introduction

Since many modern products are designed with plastics, a thorough knowledge of the mechanical behaviour of this class of material is essential. The application of accurate models to describe this behaviour will allow designers to make products lighter and cheaper. Many plastics exhibit time dependent mechanical behaviour also called viscoelasticity.

Creep is an important example of viscoelastic behaviour. Also the creep behaviour of a material is a good measure for its viscoelastic nature. In our research, an HDPE has been examined for its creep response. This material is widely used in products like containers and bottle crates and shows a strong viscoelastic behaviour.

The objective of our research is to describe the viscoelastic behaviour of HDPE with existing principles. The use of these principles such as separation of variables or time–stress superposition will stimulate the designer to apply these models.

The concept of viscoelasticity has been examined for a long period of time and it was Leaderman [1] who first was able to describe the nonlinear creep behaviour of textile fibres. For this, he introduced a nonlinear strain measure. Likewise, a nonlinear stress measure can be used:

$$\varepsilon(t) = \int_0^t D(t-\xi)\frac{\mathrm{d}(G(\sigma))}{\mathrm{d}\xi}\,\mathrm{d}\xi \tag{1}$$

This model is also called the modified superposition model [2].

This model will yield the following creep strain:

$$\varepsilon_c(t) = D(t)G(\sigma) \tag{2}$$

As can be seen this model results in separation of variables for the creep strain. The creep strain does not depend on the stress linearly but in a nonlinear way. If creep curves obeying Eq. (2) are plotted in a log time vs. log strain graph then two creep curves at two different stress levels can be superimposed by a vertical shift.

Schapery [3] introduced the time–stress superposition principle:

$$\varepsilon(t) = \int_0^t D(\psi(t)-\psi(\xi))\frac{\mathrm{d}(\sigma)}{\mathrm{d}\xi}\,\mathrm{d}\xi \tag{3}$$

where ψ denotes the reduced time:

$$\psi(t) = \int_0^t \frac{\mathrm{d}\varsigma}{a(\sigma)} \qquad \psi(\xi) = \int_0^\xi \frac{\mathrm{d}\varsigma}{a(\sigma)} \tag{4}$$

The term reduced time was first introduced by Leaderman [4] to account for the effect of temperature.

A material obeying Eqs. (3) and (4) will have the following creep strain:

$$\varepsilon_c(t) = D(t/a(\sigma))\sigma \tag{5}$$

If creep compliance curves are then plotted in a log time vs. log compliance graph then two curves at two different stresses can be superimposed by a horizontal shift. The

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Nomenclature

$a(\sigma)$	Time–stress superposition function
$D(t)$	Tensile creep compliance function
D_0	Instantaneous tensile creep compliance
$\Delta D(t)$	Transient tensile creep compliance function
$g(\sigma)$	Leaderman function
$G(\sigma)$	Leaderman function multiplied with stress
t	Time
Greek symbols	
ε	Strain
ε_c	Creep strain
σ	Stress
τ	Retardation time
ξ	Time variable
ψ	Reduced time
ζ	Time variable

nonlinear effect of a stress is a change in the time scale: at higher stress levels, the material will creep faster.

Zapas and Crissman [5] found during their research on creep and recovery of HDPE that the viscoelastic creep strain could be described by the product of a stress and time function: the Leaderman model. The stress relaxation of HDPE was studied by Popelar et al. [6]. It was found that separation of variables was again applicable, but now in terms of a strain function and a time function. The creep curves of HDPE found by Lai and Bakker [7] could be superimposed upon a master curve by shifting horizontally and vertically on double logarithmic scale.

The creep strain observed during our creep experiments on HDPE can be fitted well by a linear elastic strain and a nonlinear viscoelastic strain. The nonlinear viscoelastic strain can be found by the Leaderman model. This model is not capable of describing the behaviour just before and during yielding, i.e. at strain levels above 5%.

The nonlinear yield behaviour of ultra high modulus PE fibres was examined by Wilding and Ward [8] and it was found that the yielding can be described by two plastic Eyring [9] processes in parallel. Both plastic processes obey the principle of time–stress superposition.

Klompen and Govaert [10] made a numerical simulation of the creep behaviour of a hypothetical material with two complete viscoelastic Eyring processes in parallel. Again each process obeys the principle of time–stress superposition and consequently the yielding behaviour is identical to that found by Wilding and Ward [8]. The creep behaviour obtained by their simulation corresponds qualitatively to the results of our creep measurements.

Therefore this model with two parallel processes has been examined for its capability to describe our creep curves. It is shown that this model can describe the measured creep behaviour of HDPE accurately through yielding. Predictions with this model for constant strain rate experiments agree with experimental results.

2. Experimental

2.1. Material

HDPE 7058Z (MFI = 4.4 dg/min, $\rho = 953$ kg/m^3) is an injection moulding grade, supplied by DSM, that is suitable for crates and containers. ISO 527 3.8 mm thick tensile bars were injection moulded from this material at a relatively

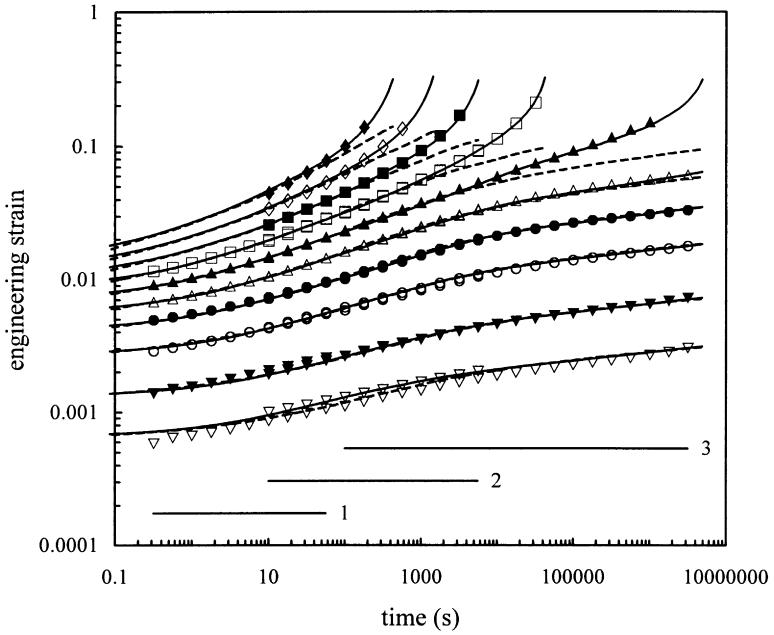

Fig. 1. Creep strain of HDPE during creep experiments at load levels of 1 (▽), 2(▼), 4(○), 6(●)…18 (◆) MPa and the Leaderman fit (dashed lines) and the two processes model fit (solid lines).

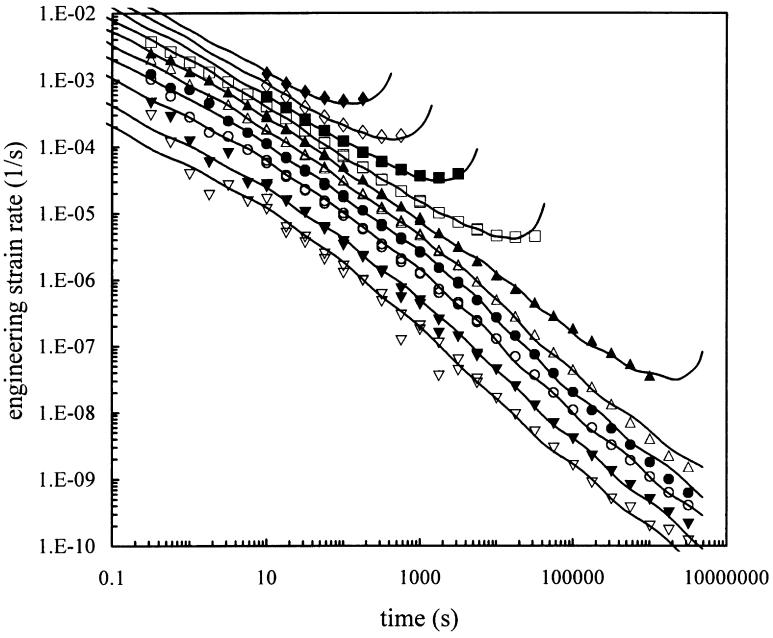

Fig. 2. Creep strain rate of HDPE during creep experiments at load levels of 1 (▽), 2(▼), 4(○), 6(●)…18 (◆) MPa and the two processes model fit (solid lines).

high nozzle temperature of 250°C and long cycle time of 55 s to minimize the effect of anisotropy and internal stresses.

It is obvious that skin and core layer will behave differently. However since the skin layer is approximately 200 μm thick [11,12], the main contribution to the mechanical behaviour will be from the nearly isotropic core layer. Therefore, creep of bars cut parallel and perpendicular to the flow direction from plates which were injection-moulded with parameters comparable to those of our tensile bars, exhibit maximum relative deviations of 7% only.

Subsequently the specimens were stored 32 weeks at 23°C and then tested at 23°C. Since all experiments were performed within ten weeks the effect of ageing can be neglected [13]. An additional test series at 43°C was performed 128 weeks after storage.

2.2. Measurements

The tensile creep measurements cover a total time period of 8 weeks. The experiments can be divided in three different categories. The average results can be seen in Figs. 1 and 2 as markers. The lines are model fits, which will be explained later.

Test series 1 was performed on a hydraulic Zwick in the Center of Polymers and Composites of the Eindhoven University. Since the constant force could be applied within 0.03 s, the measurements are reliable from 0.3 s [14]. The strain was measured with an Instron extensiometer with a gauge length of 50 mm. All measurements were carried out twice.

Test series 2 was done in the Laboratory of Mechanical Reliability at the Delft University. The servo controlled Zwick could apply the creep force in 1 second. The strain was measured with an MTS extensiometer with a gauge length of 25 mm. All tests were performed three times.

Test series 3 are dead weight experiments performed at the Laboratory of Materials Science at the Delft University. The strain was measured with an Instron extensiometer 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.

Test series 4 consists of constant strain rate experiments at strain rates of approximately 0.01, 0.001…0.000001 1/s. Measurements were carried out twice and in the same manner as in test series 2. Average results can be seen in Fig. 3 as markers. The tensile experiment at the lowest strain rate was done at an age of 64 weeks instead of 32 weeks. Tensile experiments were also performed at 43°C and a material age of 128 weeks with a wider range of strain rates. The results, to be used qualitatively only, can be seen in Figs. 4 and 5.

3. Results and discussion

In Fig. 1 it can be seen that the creep curves corresponding to stresses below about 10 MPa exhibit a pronounced convex curvature around 4000 s. This convex curvature has also been found by Turner for PP [14] and for HDPE [15]. Also the creep behaviour that Cessna [16] found for PP shows qualitatively the same behaviour as our HDPE. Struik [17] found a convex curvature in the creep curves of LDPE and also the stress relaxation curves of HDPE found by Popelar et al. [6] show a curvature (concave).

At stress levels above 10 MPa it can be seen in Fig. 2 that

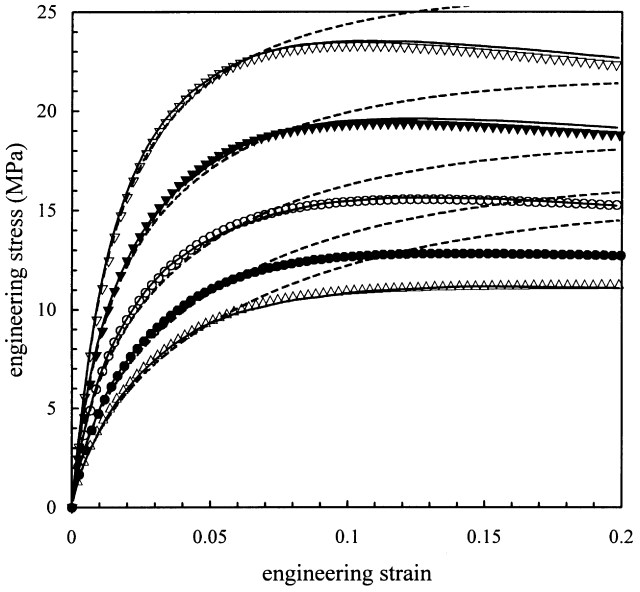

Fig. 3. Stress during straining experiments with strain rates of approximately 0.01 (▽), 0.001 (▼)…0.000001 (△) 1/s and the Leaderman predictions (dashed lines) and the two processes model predictions (solid lines).

the strain rate will start to deviate from the pattern found at low stress levels. The strain rate will decrease less, stabilize and eventually increase again, leading to an upward curvature of the creep curves in Fig. 1. This yielding of the material will lead to failure. During the 10 MPa dead weight creep experiment cracking occurred at the knife edge of the extensiometer before yielding.

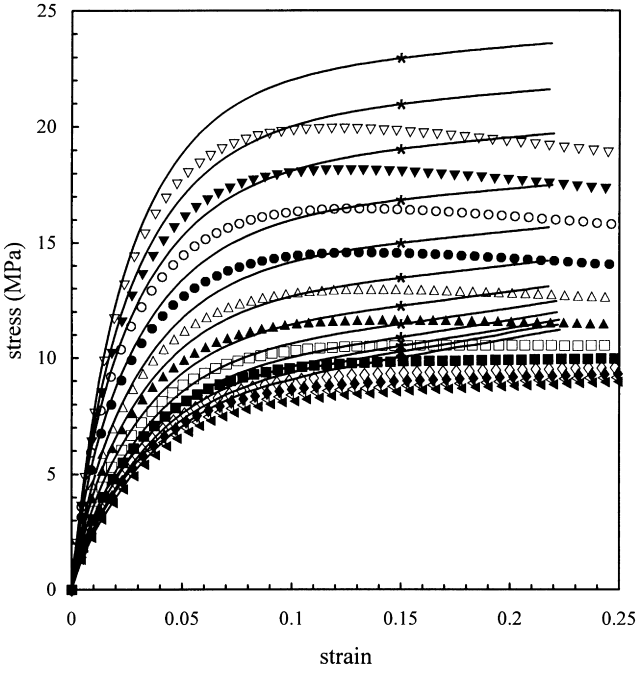

Fig. 4. Stress during straining experiments at 43°C with strain rates of approximately 0.1 (▽), 0.0316 (▼), 0.01 (○)…0.000000316 (◄) 1/s. Markers denote measurements in terms of engineering stress/strain and the lines denote the data transformed into true stress/strain.

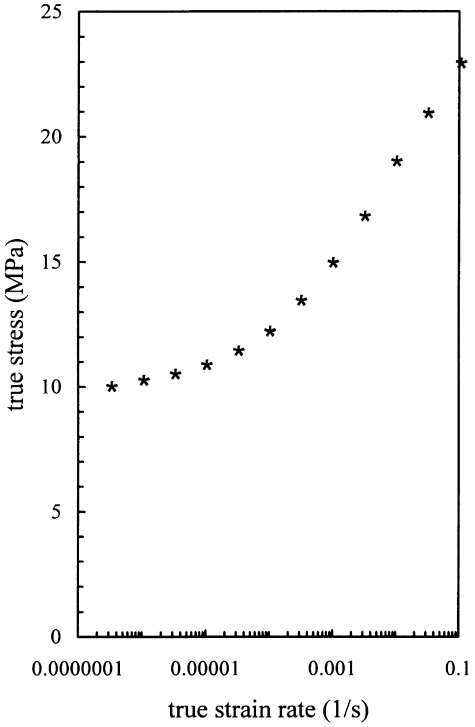

Fig. 5. True stress at a true strain level of 0.15 during straining experiments at 43°C.

The maximum strains during the measurements are just below 20%. These strain levels no longer allow engineering stresses and strains to be used. Instead, true stresses and strains must be used. The true cross-section area, needed for the true stress, is determined with the assumption of constant volume. Lai [18] 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.

3.1. Leaderman model

The occurrence of a convex curvature at a time which is independent of the stress seems to exclude the use of time–stress superposition, i.e. horizontal shifting, to describe the creep data. The Leaderman model, i.e. vertical shifting, seems more appropriate. However, the creep curves at stresses of 1–8 MPa in Fig. 1 do not superimpose exactly by a vertical shift.

Fig. 2 displays the strain rate during creep and it can be seen that these curves do superimpose with vertical shifting. This implies that the Leaderman model can be applied to the viscoelastic strain. With the assumption of linear elasticity this leads to the next creep strain:

$$\varepsilon_c(t) = D_0\sigma + \Delta D(t)\sigma g(\sigma) \tag{6}$$

where $G(\sigma) = \sigma g(\sigma)$ with $g(0) = 1$ so that the equation reduces to the linear case at low stresses.

The creep data have been fitted with this model. The $g(\sigma)$ is set equal to a fourth order polynomial and the $\Delta D(t)$ is described with the Generalized Kelvin–Voigt model [19] with retardation times of 10^i with $i \in \{-2, -1 \ldots 8\}$. Also the modulus of elasticity is a fitting parameter. Since the true stress is increasing in time, the creep strain must be determined numerically. For this a numerical integration [20] of the differential form of the Leaderman model [21] is chosen.

As can be seen in Fig. 1 the model describes the creep behaviour well up to 5% strain. The creep spectrum is shown in Fig. 6. At higher strain levels, the creep curves will exhibit an upward curvature eventually leading to yielding of the material. This can not be described by this model.

Predictions with this model for constant strain rate experiments are displayed in Fig. 3. The measured strain is given as input for the calculations. Up to approximately 7% strain, the predictions agree well with the experimental results. At higher strain levels the material starts to yield and the model is no longer valid.

3.2. Two parallel processes

For the description of the yield behaviour of many polymers two parallel Eyring processes are needed. This is not only the case for ultra high modulus PE fibres [8]. Roetling showed that this also applies for PMMA, PEMA and PP [22–24]. Also for PVC this behaviour can be observed [25]. This yield behaviour is as follows. At low strain rates only one process attributes to the total stress measured. At high strain rates, the second process starts to carry load as well. This leads to a stiffer yield behaviour at high strain rates or high yield stresses.

In Figs. 3–5 it can be seen 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. The creep curves in Fig. 1 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 MPa curve. This can also be seen in Fig. 2 where the yield points are the points with minimum strain rates. Again stiffer yield behaviour at higher stresses since here a stress increase of 2 MPa gives

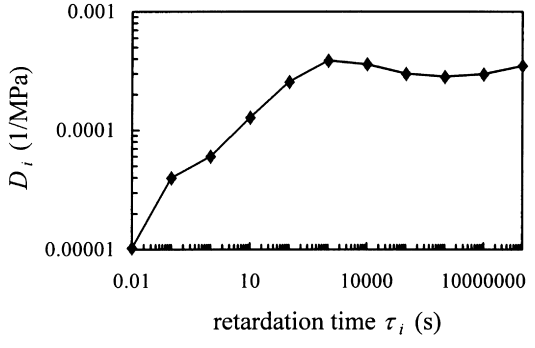

Fig. 6. Retardation spectrum (Kelvin–Viogt parameters) found by the Leaderman fit. ($D_0 = 0.000629$ 1/MPa).

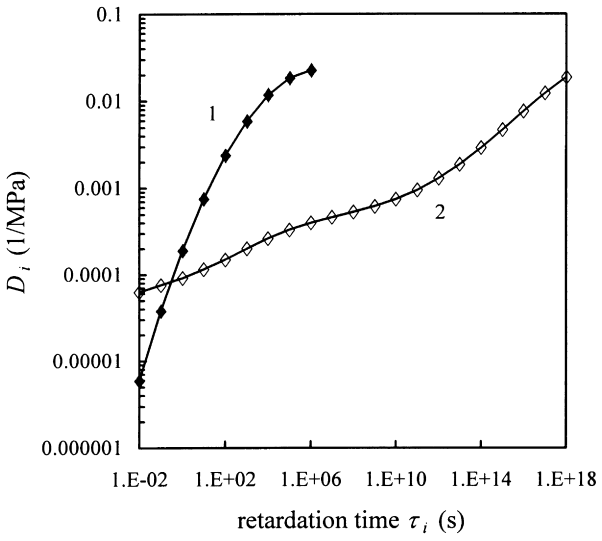

Fig. 7. Retardation spectra (Kelvin–Viogt parameters) of the two processes. ($D_{01} = 0.001394$ 1/MPa, $D_{02} = 0.001207$ 1/MPa and $\eta_1 = 2.175 \times 10^5$ MPa s).

less increase in the yield strain rate. Apparently, the yield behaviour that was measured also needs two processes to be described.

Klompen and Govaert [10] numerically simulated the creep response of a hypothetical material with two Eyring processes in parallel with each process having a complete creep spectrum. Both processes behave according to Eqs. (3) and (4) with the Eyring function for time–stress superposition:

$$a(\sigma) = \frac{\sigma/\sigma_0}{\sinh(\sigma/\sigma_0)} \tag{7}$$

The results show many similarities with our creep results. Their creep curves also exhibit a convex curvature at an almost constant time. Also the creep curves at yielding are situated closer before this convex curvature than after.

Now this model will be used to describe actual measurements. To do so two Generalized Kelvin–Voigt models [19] are placed in parallel. The true stress is equal to the sum of both true stresses while both processes have equal strains. Again, the volume is assumed to be constant for true stress determination. The response to a constant force is calculated numerically.

Process 1 is the process which only carries a load when strain rates are high. Therefore this process, which has to drop out at low strain rates, has a single (plastic) dashpot in the Generalized Kelvin–Voigt model. Process 2 does not contain a single dashpot since the tensile straining results show a continuously increasing true stress, see Fig. 4. The nominal stress shown in Figs. 3 and 4 however does decrease after its maximum.

Both spectra as well as both modulus of elasticity and the viscosity of the single dashpot in process 1 have been used to fit the creep data. Also both time–stress superposition

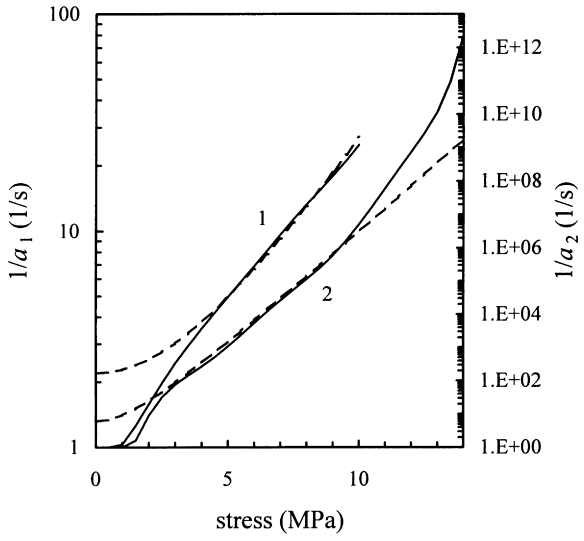

Fig. 8. Time–stress superposition functions of the two processes (solid lines) and as comparison two Eyring function approximations (dashed lines, $\sigma_{01} = 2.1$ MPa and $\sigma_{02} = 0.60$ MPa).

functions are set free during fitting and were therefore not restricted to the Eyring form (7).

The results can be seen in Fig. 1. As can be seen the fit is very good including the yield behaviour. Also the strain rate of Fig. 2 is described properly. The spectra of both processes are shown in Fig. 7. The time–stress superposition functions are displayed in Fig. 8. It can be seen that deviations from the Eyring function (7) occur at low and at high stress levels. These deviations might be the result of the difference in mechanical behaviour of the skin and the core layer.

Figs. 9 and 10 show the true stresses during creep in respectively process 1 and process 2. At short times with high strain rates, both processes contribute to the total stress. After the convex curvature, the stress in process 1 vanishes. As a result the process will enter its linear viscoelastic region. This explains why the onsets of the convex curvature are situated at the same moment in time, independent of the creep stress [10].

The vanishing of process 1 during creep may seem hard to reconcile with a convex curvature, since this will mean a

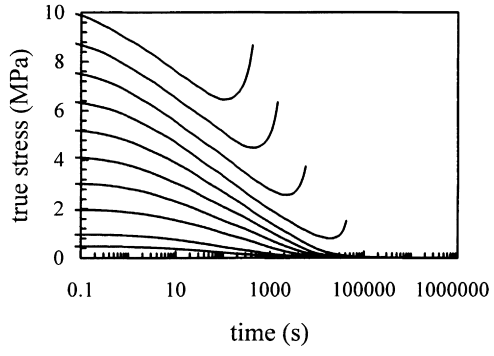

Fig. 9. True stress in process 1 during creep at 1, 2, 4, 6…18 MPa (up in graph).

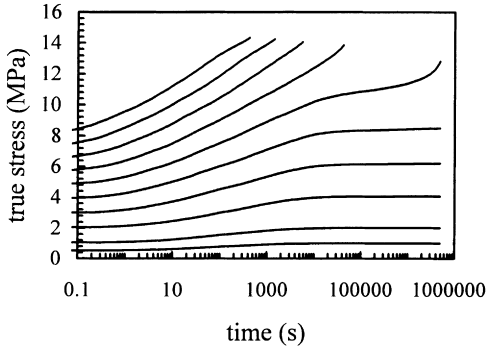

Fig. 10. True stress in process 2 during creep at 1, 2, 4, 6…18 MPa (up in graph).

decrease in creep speed. This can be understood however by consideration of a creep curve backwards in time. At first, only one process is active. When the second process is activated, the creep strain will be much less than expected from extrapolation of the one process. The result is stiffer behaviour when both processes carry load.

Predictions of the model with two processes for constant strain rate experiments are plotted in Fig. 3 together with actual data. The measurements are predicted properly over the entire range, including the yielding of the material. The model predicts very well that the faster tensile curves are further apart than the slower curves.

4. Conclusions

The Leaderman model is suitable to describe the creep behaviour of our HDPE properly as long as strains remain sufficiently below the yield point of the material; below 5% strain. Predictions of this model for constant strain rate experiments show good agreement until yielding occurs. The yield behaviour of this material can not be described with this model.

The model of Klompen and Govaert has two parallel processes with each his own time–stress superposition function. With this model the creep data can be fitted over the entire experimental range including yielding. Predictions for the constant strain rate experiments show good agreement with measurements. Also the yield behaviour is properly predicted.

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References

[1] Leaderman H. Trans Soc Rheol 1962;6:361.
[2] Findley WN, Lai JSY. Trans Soc Rheol 1967;11:361.
[3] Schapery RA. Polym Eng Sci 1969;9:295.

[4] Leaderman H. Elastic and creep properties of filamentous materials and other high polymers, Washington, DC: The Textile Foundation, 1943.
[5] Zapas LJ, Crissman JM. Polymer 1984;25:57.
[6] Popelar CF, Popelar CH, Kenner VH. Polym Engng Sci 1990;30:577.
[7] Lai J, Bakker A. Polymer 1995;36:93.
[8] Wilding MA, Ward IM. Polymer 1981;22:870.
[9] Eyring H. J Chem Phys 1936;4:283.
[10] Klompen ETJ, Govaert LE. Mech Time Dep Mat 1999;3:49.
[11] Moy FH, Kamal MR. Polym Eng Sci 1980;20:957.
[12] Fleissner M, Paschke E. Kunststoffe 1971;61:195.
[13] Struik LCE. Physical aging in amorphous and other materials, Amsterdam: Elsevier, 1978.
[14] Turner S. Polym Eng Sci 1966;6:306.
[15] Turner S. Br Plast 1964;September:19.
[16] Cessna LC. Polym Eng Sci 1971;11:211.
[17] Struik LCE. Polymer 1989;30:799.
[18] Lai J. Non-linear time-dependent deformation behaviour of high density polyethylene. PhD thesis. Delft University, 1995.
[19] Findley NF, Lai JS, Onaran K. Creep and relaxation of nonlinear viscoelastic materials, Amsterdam: NHPC, 1976.
[20] Beijer JGJ, Spoormaker, JL. Proc ANTEC'99. 1999.
[21] Czyz JA, Szyszkowski W. Comp Struct 1990;37:637.
[22] Roetling JA. Polymer 1965;6:311.
[23] Roetling JA. Polymer 1965;6:615.
[24] Roetling JA. Polymer 1966;7:303.
[25] Bauwens-Crowet C, Bauwens J-C, Homès G. J Polym Sci, Part A-2 1969;7:735.