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Tensile strength and work of fracture of oriented polyethylene fibre

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In the present investigation the influence of strain rate and temperature on modulus, strength and work of fracture of high-performance polyethylene (HPPE) fibre is studied. At low temperature and/or high strain rates the fibre shows brittle failure, displaying a pronounced strain rate and temperature dependence of the tensile strength. At high temperatures and/or low strain rates a transition from a brittle to a ductile failure mode could be observed. This brittle-to-ductile transition is analysed in terms of competitive failure modes, which leads to a simple model that can be used to predict the strain-rate dependence of the transition temperature. In the brittle failure mode it is observed that an increase in strain rate and/or decrease in temperature leads to a reduction in work of fracture. This reduction could successfully be predicted by combining a previously published mathematical model for the deformation of HPPE fibres with the observed strain-rate dependence of the tensile strength. From the numerical simulations it can be deduced that a constant minimum level for the fracture energy will be reached at high strain rates or low temperatures.

(Keywords: polyethylene fibre; tensile strength; work of fracture)

INTRODUCTION

Despite the excellent short-term mechanical properties of high-performance polyethylene (HPPE) fibres[1,2], these fibres show a pronounced time dependent behaviour under continuous static loadings. In contrast to these limiting factors, the viscoelastic character and the relatively high work-to-break of HPPE fibres compared with carbon, aramid and glass fibres renders them eminently suitable for applications where impact resistance[3–7] and vibrational damping is required[8,9]. In exploring new applications for HPPE fibres in this area, the ability to predict the influence of temperature, frequency or loading speed on the vibration damping behaviour or impact performance of these fibres and their structures is of great significance. For example, in the case of impact performance an enhanced energy absorption capacity has been reported for HPPE-fibre-reinforced composite laminates with increasing temperature[10].

Previous studies in this area were mainly directed to the deformation behaviour of polyethylene fibres and the modelling of long-term properties such as creep and stress relaxation. The deformation behaviour of melt-spun drawn polyethylene fibres has been studied extensively by Wilding and Ward[11–16]. Their model was extended to solution-spun ultra-drawn HPPE fibres by Leblans *et al.*[17] and Govaert *et al.*[18], finally yielding a mathematical model that can describe the deformation behaviour of HPPE fibres under any load and at any temperature. This model is applicable to both long loading times (creep, stress relaxation) and short loading times (dynamic excitation, impact). Using this mathematical model the dynamic and long-term properties of hybrid composites based on HPPE fibres were discussed and modelled[9,19].

The modelling referred to above includes the plastic deformation behaviour of these fibres, and is therefore capable of predicting yield stresses in constant-strain-rate experiments[18]. However, at low temperatures and/or high strain rates, the yield stress exceeds the tensile strength and the fibre will fracture before reaching the yield point. Since both the tensile strength[20] and the yield stress[18,20] depend strongly on temperature and strain rate, the failure mode is likely to change from brittle fracture to yielding with increasing temperature or decreasing strain rate[20]. Experimental observations of this change in the failure mode, further referred to as the brittle-to-ductile transition, have been reported frequently[20–23]. As the modelling published so far lacks a strain rate and temperature-dependent tensile strength, it is unable to predict work of fracture, or discriminate between the brittle and ductile failure mode.

In this study we will focus on this shortcoming and attempt to incorporate the temperature and strain-rate dependence of the tensile strength of HPPE fibres into the deformation model. This should enable us to predict the work of fracture and the brittle-to-ductile transition of these fibres.

MODELLING OF THE DEFORMATION BEHAVIOUR

Similar to the approach of Wilding and Ward[11–15] for

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melt-spun HDPE fibres, Govaert *et al.*[18] distinguished two simultaneous contributions to the deformation of solution-spun ultra-drawn polyethylene fibre:

- A recoverable or delayed elastic component, which dominates the deformation behaviour at short loading times, low stress levels or low temperatures.
- An irrecoverable or plastic component, which dominates at longer loading times, high stresses and/or temperatures.

To combine the contributions of both the delayed elastic and the plastic components in a single model, it was assumed that both components act independently and in series with each other. This leads to the following 'decomposition' of the total strain ϵ_{tot} of the fibre:

$$\epsilon_{tot} = \epsilon_{de} + \epsilon_{pl} \tag{1}$$

and therefore

$$\dot{\epsilon}_{de}(\sigma, T) = \dot{\epsilon}_{tot} - \dot{\epsilon}_{pl}(\sigma, T) \tag{2}$$

where ϵ_{de} and ϵ_{pl} are the contributions to the total strain of the delayed elastic and the plastic flow components, respectively ($\dot{\epsilon}$ is the strain rate).

To obtain a mathematical stress–strain relationship for the delayed elastic contribution, the classical theory of linear viscoelasticity is applied. The stress is related to the viscoelastic strain rate via the relaxation modulus and this relationship is represented via the Boltzmann superposition integral:

$$\sigma(t) = \int_0^t E(t - t')\dot{\epsilon}_{de}(t')\mathrm{d}t' \tag{3}$$

where σ is the extensional stress, E is the relaxation modulus and $\dot{\epsilon}_{de}$ is the strain rate acting on the delayed elastic component. To cover the viscoelastic behaviour of HPPE fibres from impact rate to long-term loadings, a continuous relaxation-time spectrum[19] for the relaxation modulus $E(t)$ can be used[9,18]. Dependent on the range of applicability, $E(t)$ can be approximated by less tortuous mathematical expressions. Leblans *et al.*[17] showed that with the introduction of a simple power-law expression for the stress relaxation modulus ($E(t) = Ct^{-n}$), the short-term linear viscoelastic behaviour of HPPE can be satisfactorily described.

The range of applicability of such a power law can be increased by using a modified power law, as follows:

$$E(t) = C(t + t_0)^{-n} \tag{4}$$

where Ct_0^{-n} is the initial modulus. This expression becomes identical to normal power-law behaviour[17] at longer loading times, whereas the continuous spectrum[18] is approximated at short loading times.

The contribution of the plastic flow to the total deformation behaviour of HPPE fibres can be characterized by long-term creep experiments and was proven to be solely determined by stress and temperature. It was shown that the stress dependence of the plateau creep rate could be described by a power-law relationship, whereas the temperature dependence could be approximated by a single thermally activated process. Stress and temperature dependence of the plastic flow rate are independent of each other and are expressed as follows:

$$\dot{\epsilon}_{pl}(\sigma, T) = a_{pl}(T)C\sigma^m \tag{5}$$

where $a_{pl}(T)$ is the temperature shift factor which incorporates the temperature dependence of the plastic flow contribution and is represented by the following expression:

$$a_{pl,T_0}(T) = \exp\left[\frac{\Delta U}{R}\left(\frac{1}{T_0} - \frac{1}{T}\right)\right] \tag{6}$$

where U is the Arrhenius activation energy, R is the gas constant, T is the absolute temperature and T_0 is the reference temperature. The substitution of equation (2) into equation (3) leads to a stress–strain relationship, which incorporates both the delayed elastic and plastic flow contributions to the total deformation of the fibre, as follows:

$$\sigma(t) = \int_0^t E(t - t')[\dot{\epsilon}_{tot}(t') - \dot{\epsilon}_{pl}(\sigma(t'))]\mathrm{d}t' \tag{7}$$

where $E(t)$ is the continuous relaxation spectrum[18], or a power law similar to equation (4), and $\dot{\epsilon}_{pl}(\sigma)$ is given by equation (5).

The validity of this relationship was demonstrated for high-speed tensile experiments as well as long-term static loadings[18]. It should be noted, however, that this equation solely predicts the stress–strain behaviour and does not provide limits to the deformation.

EXPERIMENTAL

The HPPE fibre used in this study was Dyneema™ SK60 yarn (400 denier) supplied by DSM High Performance Fibers BV.

Tensile tests on HPPE yarn with a length of 250 mm were performed on a Zwick 1445 tensile tester equipped with a thermostatically controlled oven and pneumatic fibre-clamps. Static mechanical experiments were conducted at different strain rates (10^{-5}–10^{-1} s^{-1}) and different temperatures (−40 to 80°C).

Creep experiments on HPPE yarn provided with adhesively bonded cardboard tabs were performed on a Franck 81565 tensile tester at different stress levels (250–1250 MPa) and different temperatures (23–90°C). The deformation during creep was monitored with an extensometer. Plateau creep rates were determined from the creep data employing the method suggested by Sherby and Dorn[24].

RESULTS AND DISCUSSION

In order to predict the strain-rate and temperature dependence of the energy absorption of HPPE fibres, the deformation behaviour and strength of the fibres as a function of time and temperature should first be characterized.

Characterization of fibre deformation

Recoverable deformation. To a first approximation, HPPE fibres can be regarded to be linear viscoelastic in dynamic and/or short-term loading situations, as encountered in impact[9,18]. Well known characterization methods for linear viscoelastic materials, which cover a sufficiently wide time–frequency range, are dynamic excitations, creep or stress relaxation experiments. Dynamic excitation, for example, was used in a previous

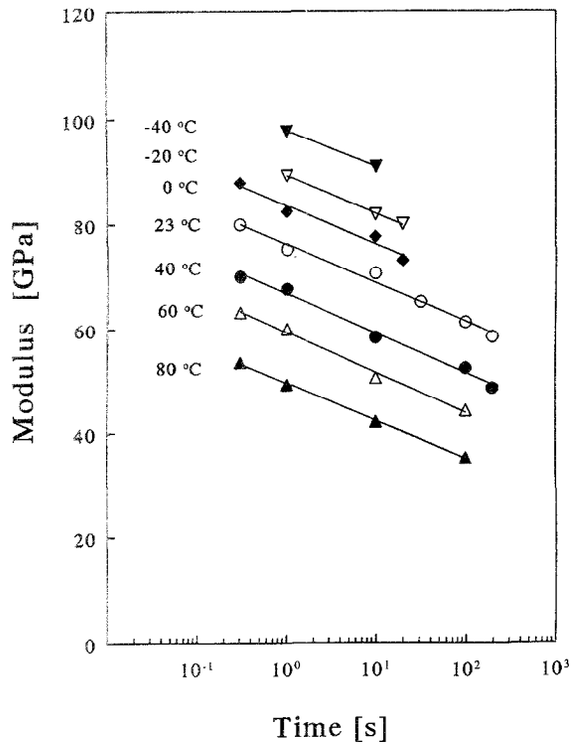

Figure 1 Relaxation modulus of SK60 fibre as determined from tensile tests vs. time at temperatures from −40 to 80°C

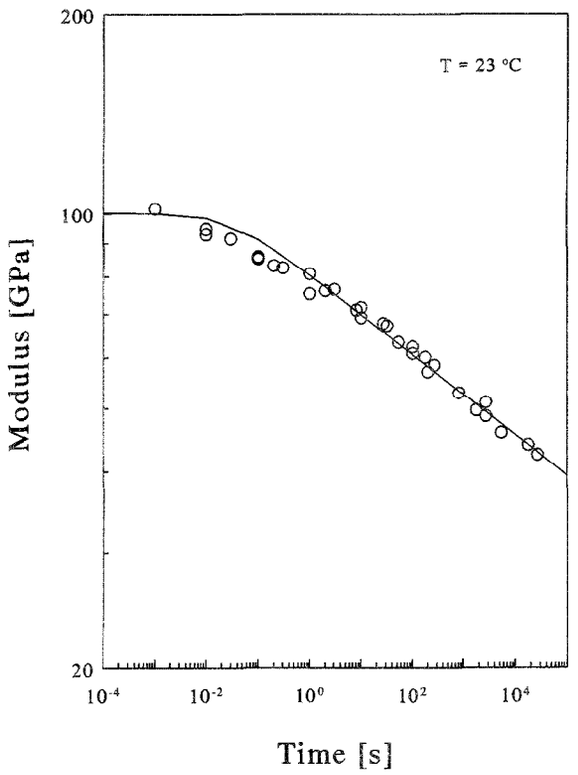

Figure 2 Master curve of the modulus of SK60 fibre at room temperature as a function of loading time; solid line gives the model prediction using the modified power law (equation (4))

study for the characterization of the dynamic modulus at different temperatures and frequencies[9,18]. In this present study, an alternative approach using conventional tensile tests at different strain rates and temperatures has been followed in order to include the determination of fibre strength. In the case of linear viscoelastic behaviour, it can be shown that the following relationship applies:

$$\frac{\partial \sigma}{\partial \epsilon} = E\left(t = \frac{\epsilon}{\dot{\epsilon}}\right) \tag{8}$$

Therefore, values for the relaxation modulus $E(t)$ can simply be determined from tangent moduli at a fixed strain level. To cover a sufficiently wide time–frequency range, tensile experiments were performed at strain rates which varied from 10^{-5} to $10^{-1}\,s^{-1}$ and temperatures ranging from −40 to 80°C. From all of these stress–strain curves tangent moduli at 1% strain were derived and subsequently transferred into isotherms for the stress relaxation modulus (*Figure 1*).

A master curve for the modulus at 23°C, constructed by horizontal shifting and matching of these isotherms, is presented in *Figure 2*. Since time–temperature superposition is valid for HPPE[18], this curve can be used to predict the actual fibre behaviour over a large time-scale. The temperature dependence can be described by a thermally activated Arrhenius process with an activation energy of $115\,kJ\,mol^{-1}$.

The time dependence was fitted using the modified power law given in equation (4), where $C = 80.54\,GPa\,s^{-n}$, $n = 0.062$ and $t_0 = 0.03\,s$.

Irrecoverable deformation. Since the plastic-flow component dominates the deformation of the fibre at long loading times, creep experiments are eminently suitable for characterizing this contribution to the deformation. At high temperatures, additional information on the stress dependence of the plastic-flow contribution can be obtained in deriving yield stresses from constant-strain-rate experiments[15,18]. The stress dependence of the plateau creep rate, as measured for SK60 fibres in creep experiments, is presented for various temperatures in *Figure 3*. It is shown that, similar to observations for a different type of HPPE fibre (Dyneema SK66)[18], the stress dependence of the plateau creep rate can be represented by a power-law relationship at all temperatures and that the slope of these power laws is independent of temperature. Consequently, the stress and temperature dependence of the plastic flow rate can be described by using equations (5) and (6), which leads to the following:

$$\dot{\epsilon} = C_{pl}\sigma^m \tag{9}$$

where

$$C_{pl} = 0.22 \times 10^{12} \exp\left(-12\,300 T^{-1}\right)\,(GPa^{-m}\,s^{-1})$$

and $m = 4$

An expression for the yield stress σ_y can be obtained by inversion of equation (9), which leads to the following:

$$\sigma_y = \left(\frac{\dot{\epsilon}}{C_{pl}}\right)^{1/m} \tag{10}$$

The temperature dependence of the irrecoverable creep could be approximated by a single temperature-activated process with an activation energy of $120\,kJ\,mol^{-1}$. This value is identical to the activation energy of the dielectrical α-relaxation[25], which is a strong indication that the underlying molecular mechanism is

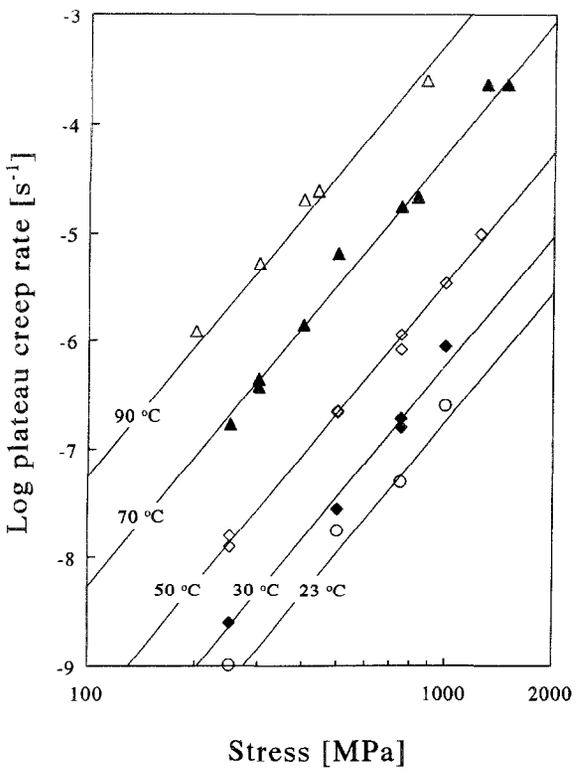

Figure 3 Stress dependence of the plateau creep rate of SK60 fibre at temperatures from 23 to 90°C

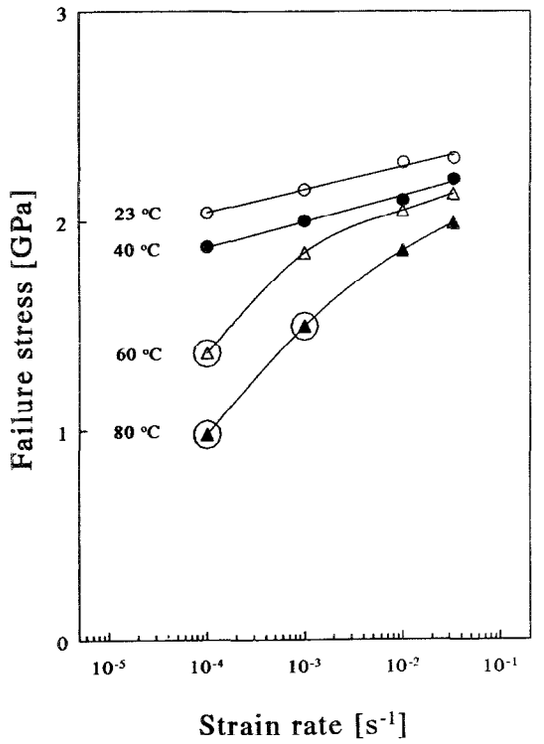

Figure 4 Failure stress as a function of strain rate of SK60 fibre at temperatures ranging from 23 to 80°C, circled data points indicate a ductile failure mode (yielding)

the diffusion of *gauche* defects within the crystalline regions[26]. Since the diffusion of such defects results in a 180° rotational-C/2 translational movement of the crystalline stem throughout the crystalline lattice[27,28], this motion can easily induce chain slip under the influence of an externally applied stress. This interpretation is in full accordance with the results and conclusions of Wilding and Ward in the case of melt-spun/drawn HDPE fibres[12,15].

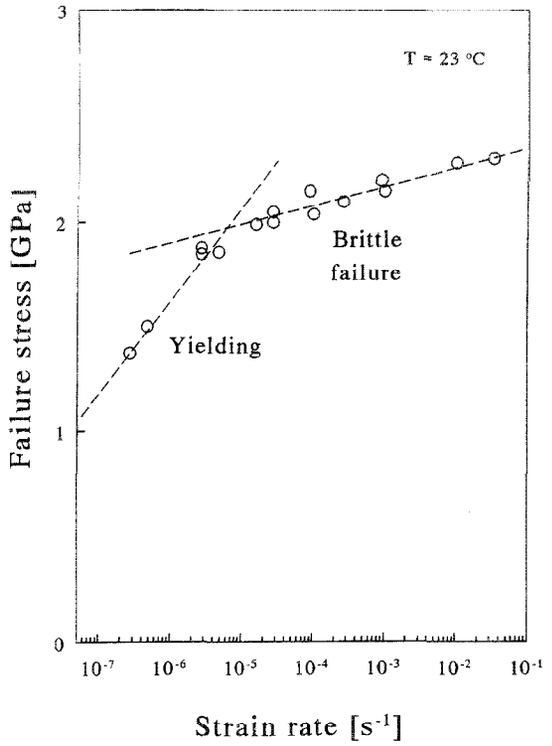

Figure 5 Mastercurve for fibre strength of SK60 fibre at room temperature

Characterization of fibre strength

Isotherms for the failure stress of HPPE yarn are constructed from tensile experiments at different temperatures and strain rates. From *Figure 4* it can be observed that an increase in temperature leads to a decrease in strength or yield stress. Yielding was observed at extremely low strain rates and/or high temperatures. These results are in full accordance with other studies on the influence of strain rate[15,20,29–31] and temperature[21–23] on the strength of HPPE fibres, showing a decrease in failure stress and a transition from (pseudo) brittle fracture to yielding with decreasing strain rates and/or increasing temperatures.

A master curve for the failure stress of HPPE fibres at 23°C is constructed by horizontal shifting of the isotherms (*Figure 5*). From this master curve it can be concluded that even at room temperature, yielding will occur at sufficiently low strain rates ($< 4 \times 10^{-6}$ s^{-1}).

The relationship between strain rate and the failure stress in the brittle region, further referred to as the tensile strength, could be excellently described with a power-law relationship similar to equation (10), as follows:

$$\sigma_b = \left(\frac{\dot{\epsilon}}{C_{br}}\right)^{1/n} \tag{11}$$

where

$$C_{br} = 1.95 \times 10^{5} \exp(-9494T^{-1}) \text{ (GPa}^{-n}\text{ s}^{-1}\text{)},$$

and $n = 48$

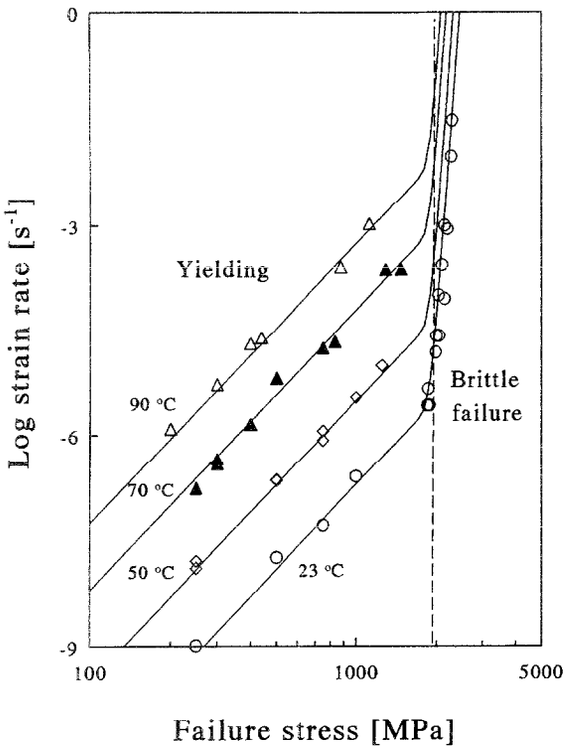

Figure 6 Strain rate as a function of failure stress of SK60 fibre showing a clear transition from yielding to brittle fibre fracture

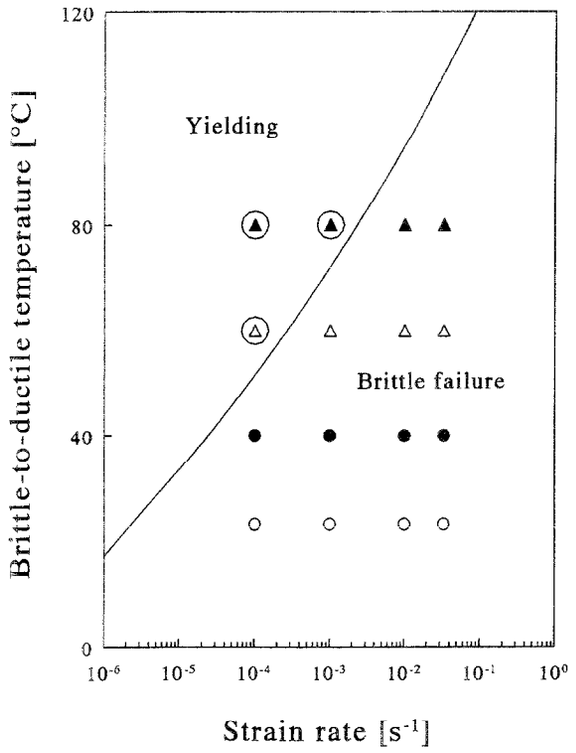

Figure 7 Brittle-to-ductile transition temperature as a function of applied strain rate for SK60 fibre, including experimental data taken from *Figure 4*: solid line represents an analytical prediction based on equations (10) and (11); circled data points indicate a ductile failure mode (yielding)

Also in this case, the temperature shift can be described by one thermally activated process with an activation energy of approximately 85 kJ mol^{-1}, which is comparable to that of the reversible deformation process. Similar values for the fracture process of ultra-high-molecule-weight PE fibres were also reported by Smook *et al.*[29]. Since these values for the activation energy are significantly lower than the bond energy of a C–C bond in polyethylene ($\sim$ 300 kJ mol^{-1}), these data support previous studies which indicated that the failure process of HPPE fibres for a large extent is governed by chain slippage, i.e. breakage of secondary bonds[29,31,32].

The brittle-to-ductile transition

The results of the long-term creep experiments and the tensile experiments can be combined to describe the failure or yield stress of the HPPE fibre over large regions of strain rates. In this approach the failure stress is interpreted in terms of a competition between yielding and brittle fracture (i.e. the lowest value determines the failure mode).

Figure 6 presents the competition between both failure modes in a double logarithmic plot of strain rates *versus* failure stress, combining equations (9) and (11). The transition from ductile-to-brittle failure can be observed as a pronounced change in the slope of the curve.

The strain-rate dependence of the brittle-to-ductile transition temperature is plotted in *Figure 7*. This plot was constructed by combining equations (10) and (11), assuming that the yield stress equals the tensile strength at the brittle-to-ductile transition. From the resulting expression a relationship between the brittle-to-ductile transition temperature and the applied strain rate could be derived, which is represented in *Figure 7* as a solid line. Above this line the tensile strength exceeds the yield stress, implying a ductile failure mode. Below this line the situation is reversed and brittle fracture is observed. For further illustration the experimental data of *Figure 4* have been included in *Figure 7*.

The temperature dependence is often evaluated in a plot of failure stress *versus* temperature. This plot is presented in *Figure 8* for three different strain rates and was constructed by combining equations (10) and (11). From *Figure 8* two important observations can be made: (i) the brittle-to-ductile transition temperature is highly strain-rate dependent, and (ii) the temperature dependence of the yield stress is strongly non-linear, becoming less pronounced at higher temperatures.

These observations put a new perspective on the conclusions from Dijkstra *et al.*[21], van de Werff and Pennings[22] and Dessain *et al.*[23], who measured the failure stress of a highly drawn polyethylene fibre as a function of temperature at a constant strain rate. By linear extrapolation it is concluded that the yield stress decreases to zero at 152°C. Based on this extrapolation the molecular mechanism of flow in oriented polyethylene is assigned to the transition from the orthorhombic crystalline phase into the hexagonal phase, a solid–solid transition which is known to occur at 152°C in constrained polyethylene fibres[33]. Since it is clear from *Figure 7* that even a small change in strain rate would drastically influence the intersection point with the temperature axis, these present authors strongly question such an interpretation.

Work of fracture

Since the stress–strain behaviour and the failure stress of the fibre are now fully characterized, work of fracture

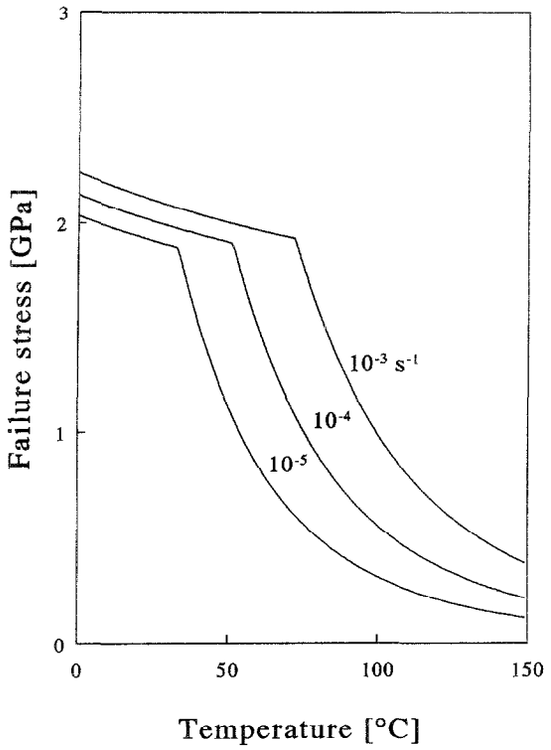

Figure 8 Failure stress as a function of temperature of SK60 fibre for various strain rates

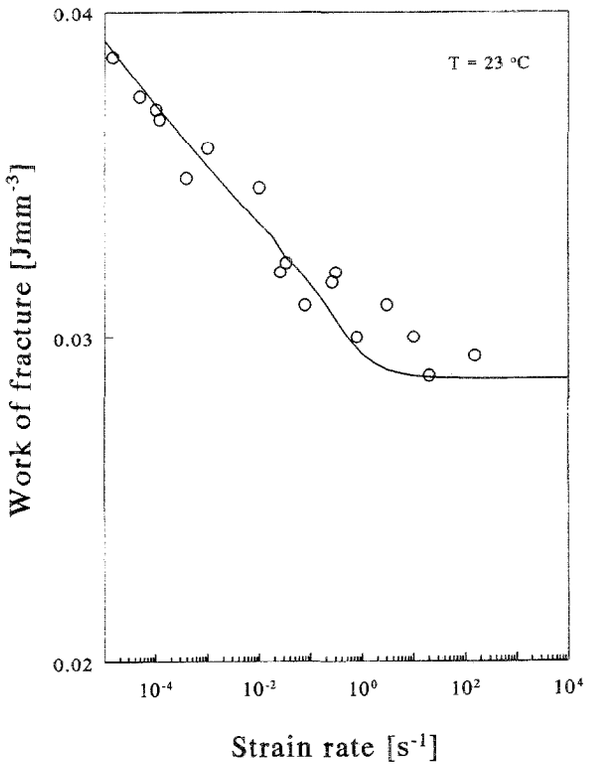

Figure 9 Specific work of fracture *versus* strain rate for SK60 fibres: (O) experiment; (—) model prediction

can be predicted by using the following expression:

$$U(\epsilon) = \int_0^{\epsilon_0} \sigma(\epsilon) \mathrm{d}\epsilon \qquad (12)$$

where ϵ_b is the failure strain and $\sigma(\epsilon)$ can be calculated for different strain rates using equation (7), since both the relaxation modulus $E(t)$, (equation (4)), and the plastic-flow contribution (equation (5)) are known. The strain at break can be predicted by combining the predictions for $\sigma(\epsilon)$ and the strain-rate dependence of the failure stress in the brittle region (equation (11)). The work of fracture can subsequently be determined by simple integration of the area under the predicted stress–strain curve. The predicted work of fracture at room temperature is presented as a function of strain rate in *Figure 9*. The work of fracture of the HPPE fibres is calculated from the experimental data by integration of the area under the stress–strain curves. To facilitate comparison of the experimental data with the model predictions, the isotherms of work of fracture were shifted to a master curve at room temperature. The work of fracture for HPPE fibres decreases with increasing strain rate and results in a constant minimum fracture energy of $\sim 0.03\,\mathrm{J\,mm^{-3}}$. Numerical predictions are in good agreement with experimental data, showing the validity of the model.

From the graph in *Figure 9* it can be concluded that the work of fracture of HPPE fibres increases with decreasing strain rate and/or increasing temperature. This observation puts the temperature dependence of HPPE fibres and composites in an interesting perspective. On the one hand structural properties such as stiffness and strength decrease with an increase in temperature, whereas on the other hand energy absorption or impact resistance, being the most outstanding property of structures based on HPPE fibres, improves due to an increase in failure strain with an increase in temperature. In a separate study this mathematical model for the work of fracture of HPPE fibre was therefore also used to model the impact behaviour of HPPE-fibre-reinforced composites by quantitatively describing the contribution of fibre fracture to the total energy absorption during penetration of a composite laminate[34].

CONCLUSIONS

The strain rate and temperature dependence of the tensile strength of HPPE fibre was evaluated. By assuming a competition between a brittle and a ductile (yielding) failure mode the strain-rate dependence of the brittle-to-ductile transition temperature could be predicted. It was shown that the brittle-to-ductile transition temperature shifts to higher values with increasing strain rate.

By incorporating the strain-rate dependence of the tensile strength in an existing model for the stress-strain behaviour of HPPE fibres, a mathematical evaluation of the work of fracture could be performed. Numerical predictions of the work of fracture of HPPE fibres in the brittle region are in excellent agreement with experimental observations. The results indicate that the work of fracture decreases with decreasing temperature and/or increasing strain rate, reaching a constant minimum value of $0.03\,\mathrm{J\,mm^{-3}}$ at high strain rates.

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