JOURNAL OF MATERIALS SCIENCE 16 (1981) 389–396

Creep behaviour of polyethylene and polypropylene

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Tensile creep tests and stress reduction studies during creep have been carried out for polyethylene and polypropylene. The results obtained suggest that a consistent approach for the presentation of creep data for these polymeric materials can be obtained since the creep curves at 293 K for polyethylene and polypropylene over a wide stress range can be superimposed by describing the variation of creep strain, ϵ, with time, t, as $\epsilon = \epsilon_0 + \epsilon_p[1 - \exp(-K\dot{\epsilon}_s t)] + \dot{\epsilon}_s t$, where ϵ_0 is the initial strain on loading, ϵ_p is the primary creep strain, $\dot{\epsilon}_s$ is the secondary creep rate, and K is a constant.

1. Introduction

The creep properties exhibited by thermoplastics are determined not only by the stress, temperature and humidity conditions during testing but also by the molecular structure, which is affected by the conditions under which the samples have been processed and stored [1]. The approaches used to study creep of polymers vary widely both in the testing procedures and in the methods of presenting creep data [2, 3]. Several studies have also considered the response to stress reductions during creep and the consequent recovery of creep strain [4, 5]. Since a detailed understanding of creep and recovery behaviour is needed for optimum exploitation of thermoplastics as engineering materials, the present programme was designed to investigate the creep characteristics of polyethylene and polypropylene by examination of (a) the form of the creep curves displayed, (b) the stress dependence of the creep parameters and (c) the strain–time response following various stress reductions during creep.

2. Presentation of creep data

High density polyethylene and polypropylene were obtained in the form of compression moulded sheets of approximately 2.54 mm thickness. Test-pieces having a gauge length of 31.75 mm and a width of 6.35 mm were cut from these sheets and tested in tension using a constant load creep machine with a 2:1 lever loading ratio. The specimen was held in friction grips having a screw thread through the centre of the specimen ends. Under uniaxial tension, the creep lateral contraction ratio, $\nu_c(t)$, can be defined as the ratio of the lateral strain to the axial strain recorded after a fixed time, t. Although it has generally been found that the time and stress dependence of the creep lateral contraction ratio is low compared with the axial strain behaviour during tensile creep [6, 7], only the axial creep strain was monitored in the present programme by means of a light aluminium extensometer clamped to the ends of the specimen gauge length. The extensometer incorporated a pair of differential capacitance transducers capable of measuring changes in specimen length of 10 nm. The creep machine was located in a constant temperature room which was maintained at 293 (± 0.25) K.

The variation of the creep strain with time is often presented as a semi-logarithmic plot as illustrated in Fig. 1. However, with this approach, the change in creep rate with time is not immediately apparent. With the present materials, the high initial creep rate recorded immediately after the initial extension on loading decreased continuously with time to an apparently constant rate (Fig. 2). Eventually, this constant rate period was followed by a stage of gradually increasing creep rate which commenced when the strain accumu-

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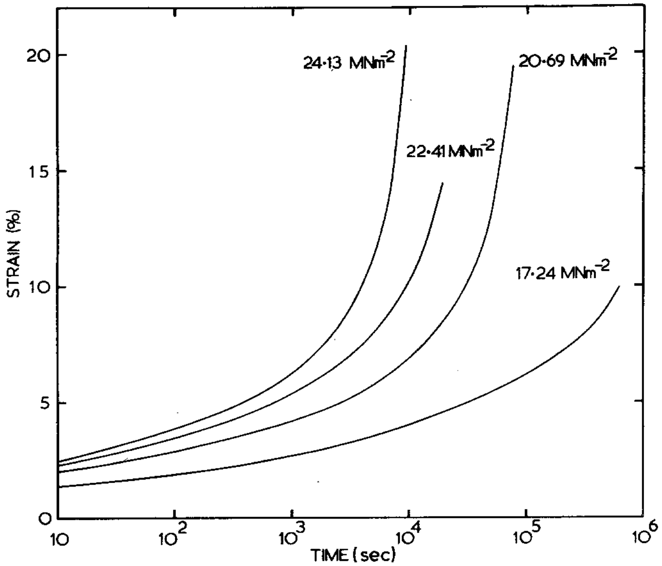

Figure 1 Creep curves obtained for polypropylene over a range of stresses at 293 K, plotted with a logarithmic time scale. The tests were discontinued at the times indicated.

lated in a highly localized manner through the occurrence of mechanical instability causing a neck to form in the specimen gauge length [8, 9].

The variation of the creep strain, ϵ, with time, t, for rigid plastics such as high density polyethylene [10], polyvinyl chloride [11] and cellulose nitrate [12] has been represented by an equation of the form

$$\epsilon = \epsilon_0 + \beta t^p, \qquad (1)$$

where ϵ_0 is the initial specimen extension on loading and β and p are constants (with $0 < p < 1$). However, equations of this form were not found to satisfactorily describe the data obtained in the present work. It can be seen from Fig. 3 that for all stress levels examined, a definite steady-state creep rate was established, showing a constant creep rate over a strain range of ~ 0.04. Thus, the creep behaviour cannot be described by equations which predict continuously decreasing creep rates (Equation 1) or points of inflection [13]. Moreover, Equation 1 predicts an infinite initial creep rate on loading (at $t = 0$), whereas finite values were invariably observed.

An equation which leads to a constant secondary creep rate and does not predict an infinite initial creep rate can be derived by assuming that a single mechanism controls the primary and secondary creep behaviour and that the material obeys the laws of unimolecular kinetics [14]. The rate constant can be expressed as $K\dot{\epsilon}_s$ where $\dot{\epsilon}_s$ is the

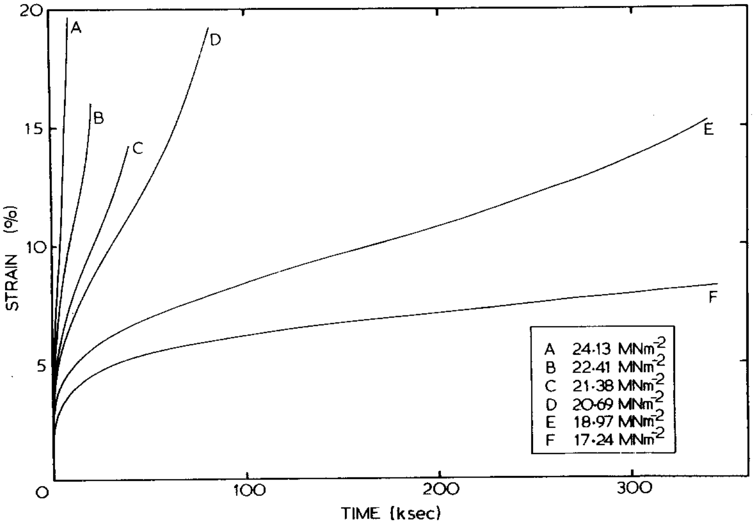

Figure 2 Strain/time relationships for polypropylene tested at different stress levels at 293 K.

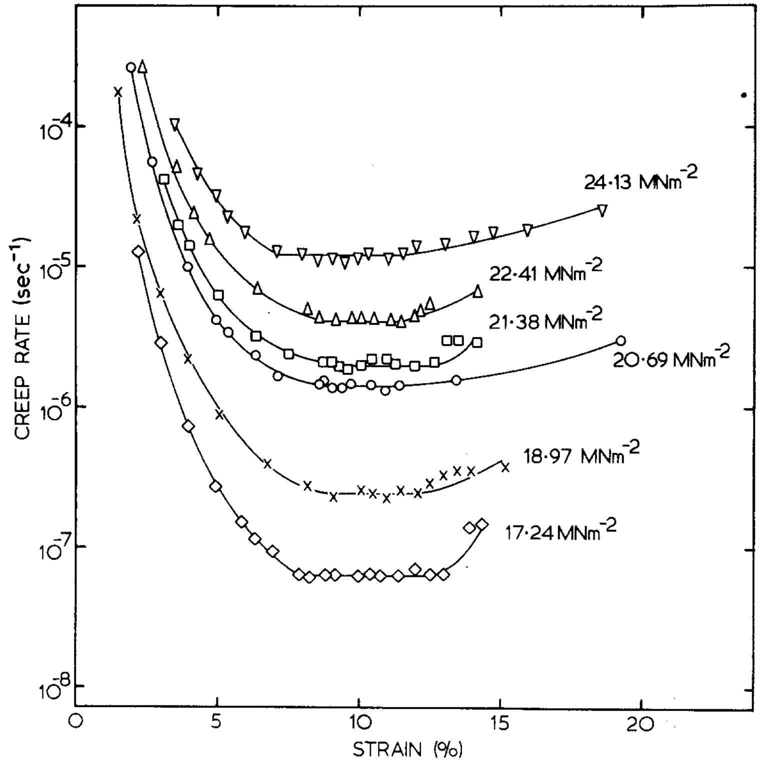

Figure 3 The variation of the creep rate with strain for polypropylene tested at various stress levels at 293 K.

secondary creep rate and K is a constant [15] as below.

$$\frac{\mathrm{d}}{\mathrm{d}t}(\dot{\epsilon}-\dot{\epsilon}_s) = -K\dot{\epsilon}_s(\dot{\epsilon}-\dot{\epsilon}_s). \qquad (2)$$

Integration of this expression gives

$$\epsilon = \epsilon_0 + \left(\frac{\dot{\epsilon}_0 - \dot{\epsilon}_s}{K\dot{\epsilon}_s}\right)[1-\exp(-K\dot{\epsilon}_s t)] + \dot{\epsilon}_s t, \qquad (3)$$

where ϵ_0 is the instantaneous strain on loading and $\dot{\epsilon}_0$ is the initial creep rate when $t = 0$. Furthermore, if the same process determines the primary and secondary creep rates then $\dot{\epsilon}_0 = A\dot{\epsilon}_s$ so that the total primary strain, ϵ_p, is given by [15]

$$\epsilon_p = \left(\frac{\dot{\epsilon}_0 - \dot{\epsilon}_s}{K\dot{\epsilon}_s}\right) = \left(\frac{A-1}{K}\right). \qquad (4)$$

It has previously been established that the creep curves recorded for polyvinyl chloride [15] can be described using Equation 3. To examine the applicability of this approach, the values of the parameters in Equation 3 were derived by a least squares regression analysis for each test carried out in the present programme. The accuracy with which the equation describes the creep curves was then determined by computing the difference between the measured creep strains and the theoretical values given by Equation 3 using the derived constants. Throughout most of the primary and secondary creep periods (up to a total creep strain of ~ 0.12) the average difference between the experimental strains, ϵ, and the calculated values, ϵ_c, was < 0.0002. However, during the initial 10 to 20% of the primary creep life, the difference $(\epsilon_c - \epsilon)$ increased progressively as t approached zero, i.e. the actual creep rate decreased more rapidly than is predicted from Equation 3 (Fig. 4). Although a deviation from Equation 3 at the start of the creep tests was not reported for polyvinyl chloride [15], the behaviour shown in Fig. 4 was invariably found for the creep tests carried out with polyethylene and polypropylene. Even so, the present results show that the creep curves recorded for these materials can be accurately described by Equation 3 over most of the primary and secondary creep lives.

3. Stress dependence of creep parameters

The creep properties of thermoplastics can vary markedly with stress, i.e. these materials behave in a non-linear viscoelastic manner. Creep tests were therefore carried out for both polyethylene and polypropylene over a range of stresses at 293 K. The stress dependence of the secondary creep rate can be represented by an equation

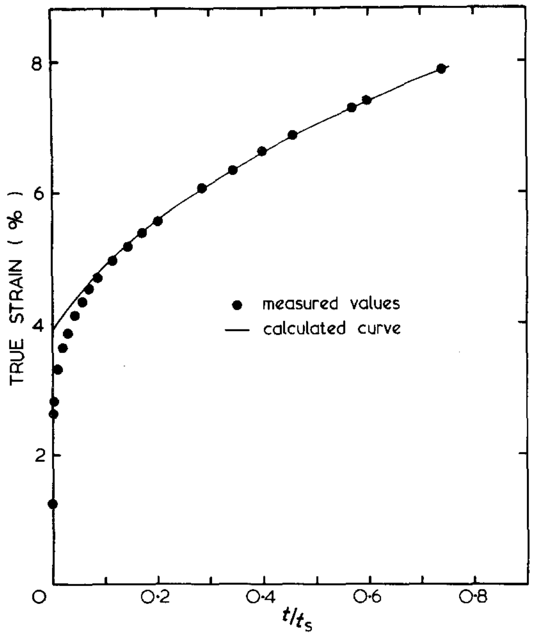

Figure 4 The variation of the true creep strain with time during primary creep of polypropylene at 20.69 MNm^{-2} and 293 K. The time to the onset of steady state creep, t_s, was 21 ksec.

of the form

$$\dot{\epsilon}_s \alpha \sigma^n \tag{5}$$

with $n \simeq 11$ for polyethylene and $\simeq 16$ for polypropylene (Fig. 5). In addition, a further series of tests was carried out for polyethylene at 293 K in which specimens were tested in tension using an Instron machine over a range of strain rates comparable with those found in the creep experiments. For each tensile test, the stress required to maintain a given constant strain rate increased until a maximum value was attained, after which the stress level decreased gradually as necking occurred. Measurements of the strain rate sensitivity of the maximum stress are included in Fig. 5. Clearly, the results obtained are directly comparable with those obtained for creep, showing that the stress dependence of the deformation rates can be described by Equation 5, independent of the method of testing.

To examine the suggestion that the processes controlling the creep behaviour of polyethylene and polypropylene are the same throughout primary and secondary creep, a study was made to determine whether the stress dependence of the primary creep parameters was similar to that for the secondary creep rate. From Equation 4, the primary creep parameters are related to $\dot{\epsilon}_s$ since, at $t = 0$,

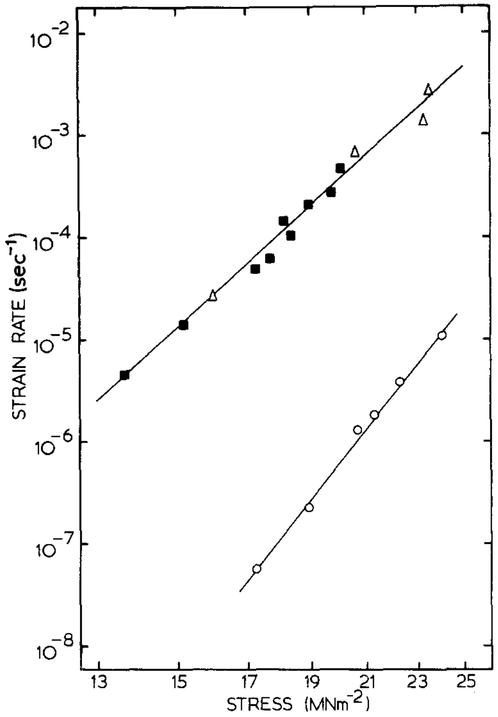

Figure 5 The dependence of the steady state creep rate, $\dot{\epsilon}_s$, on the applied stress, σ, for polypropylene (○) and polyethylene (■) at 293 K. Also shown is the strain rate dependence of the maximum stress recorded in tensile tests carried out for polyethylene (△) at 293 K.

$$\dot{\epsilon}_0 = (K\epsilon_p + 1)\dot{\epsilon}_s, \tag{6}$$

where $\dot{\epsilon}_0$ is the computed initial creep rate using the derived constants for each creep test. It was found that the ratio $\dot{\epsilon}_0/\dot{\epsilon}_s$ was constant (~ 4) over the entire stress range employed. Yet, as a result of the deviation from Equation 3 at the start of the creep test (Fig. 4), the computed values of $\dot{\epsilon}_0$ are considerably lower than the measured initial creep rates, $\dot{\epsilon}_i$, (Fig. 6). Even so, the stress dependences of both $\dot{\epsilon}_0$ and $\dot{\epsilon}_i$ are the same as that for $\dot{\epsilon}_s$.

When the primary and secondary creep properties can be represented by Equation 3, the primary stage ends when $\epsilon_p\ [1 - \exp(-K\dot{\epsilon}_s t)]$ becomes indistinguishable from ϵ_p. Measurements of the time to the onset of the secondary stage, t_s, established that

$$K\dot{\epsilon}_s t_s \simeq 4. \tag{7}$$

Thus, with the present accuracy of strain measurement, the primary stage appears to be complete when $[\exp(-K\dot{\epsilon}_s t)] \simeq 0.02$. On this basis, the creep rate at any fraction of the primary stage

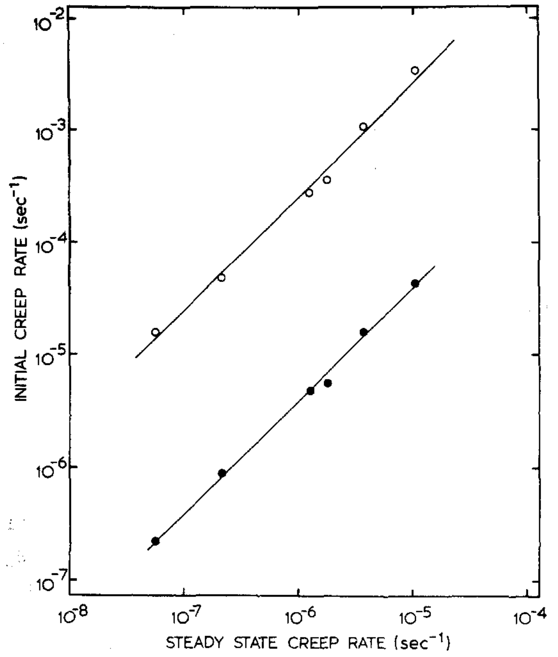

Figure 6 The relationship between the steady state creep rate, $\dot{\epsilon}_s$, and the initial creep rates for polypropylene. Open symbols represent measured values ($\dot{\epsilon}_i$) and closed symbols are calculated values ($\dot{\epsilon}_0$).

displayed at different stress levels is directly proportional to the secondary creep rate, confirming that the same mechanism controls the creep behaviour of polyethylene and polypropylene up to the onset of neck formation.

Although general agreement has not been reached on a precise description of the creep behaviour of thermoplastics [7], the present results support the view [15] that a consistent approach can be developed through Equation 3. Using the values of the creep parameters derived for polypropylene gives

$$(\epsilon - \epsilon_0) = 0.0178[1 - \exp(-166\dot{\epsilon}_s t)] + \dot{\epsilon}_s t. \quad (8)$$

Similarly, for polyethylene, the equation takes the form

$$(\epsilon - \epsilon_0) = 0.0301[1 - \exp(-97\dot{\epsilon}_s t)] + \dot{\epsilon}_s t. \quad (9)$$

The effects of variations in testing conditions are therefore contained in this equation through the dependence of the secondary creep rate on stress, temperature, etc. The validity of this approach is then illustrated by the observation that the creep curves recorded at different stress levels for polypropylene can be superimposed* on the theoretical curve given by Equation 8 (Fig. 7).

4. Recovery of creep strain following stress reductions

Considerable attention has been paid to the study of the recovery of creep strain following complete removal of the load during creep of polymeric materials [2, 16]. In contrast, relatively few studies have been made of the strain–time behaviour which occurs after removal of only part of the load [16]. In the present programme, polypropylene was used to examine the recovery

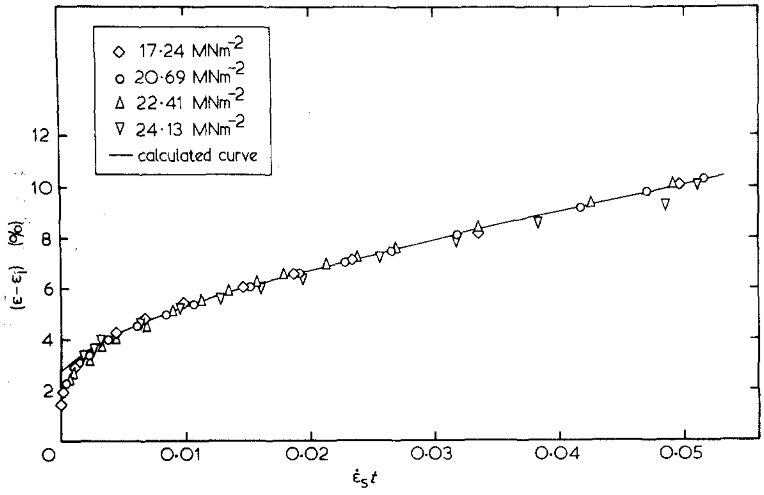

Figure 7 The creep strain/time curves obtained at different stress levels for polypropylene at 293 K superimposed by rationalizing the time scales through the steady creep rates, $\dot{\epsilon}_s$, recorded for each test.

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*The deviation of the experimental results from Equation 8 at the beginning of the creep tests means that the computed value of the initial extension on loading, ϵ_0, is larger than the value actually obtained, ϵ_i. Thus, in superimposing the theoretical line given by Equation 8 on the actual values obtained, a correction for this difference has to be made (Fig. 7).

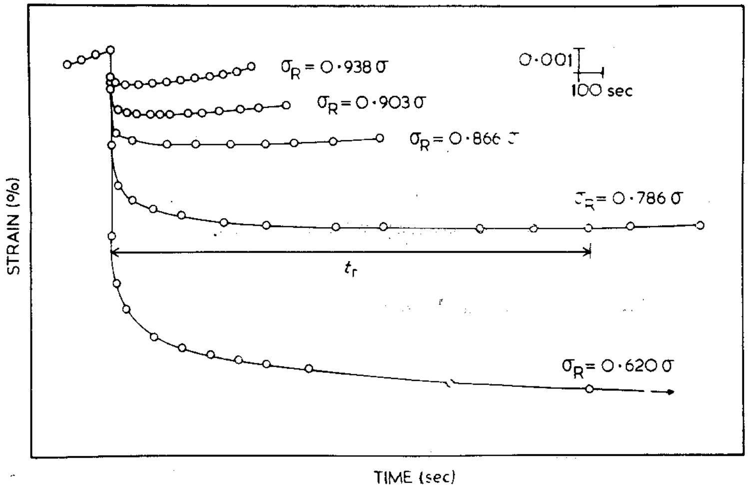

Figure 8 The strain–time behaviour observed when the applied stress, σ, was reduced during secondary creep of polypropylene at 24.13 MNm^{-2} and 293 K to continue the test at a reduced stress level, σ_R.

of creep strain which took place when the applied stress, σ, was reduced by varying amounts, $\Delta\sigma$ (in the range 0.05 to 0.54 σ) during secondary creep, and the test continued at the lower stress level, $\sigma_R (= \sigma - \Delta\sigma)$. Each individual stress reduction was found to be accompanied by an instantaneous specimen contraction and invariably followed by a period of recovery in which the negative creep rate observed decayed to zero before a new positive rate was eventually established (Fig. 8). Clearly, the magnitude and duration of the period of negative creep increased and the final positive creep rate decreased with larger stress reductions.

In load removal studies during creep of polycarbonate [17], it was reported that the instantaneous specimen contraction on decreasing the load to zero was found to exceed the initial extension obtained on loading at the start of the creep test. In the present work on polypropylene, two instantaneous strain measurements were made from the stress change experiments; namely, ϵ_u, the instantaneous contraction obtained immediately on reducing the load by $\Delta\sigma$, and ϵ_L, the immediate specimen extension observed on reloading back to the original stress level after the positive creep rate was attained under the reduced stress. These values, together with the instantaneous extension, ϵ_i, recorded on loading at the commencement of the creep tests, are presented in Fig. 9. It must be noted that the very fast creep rates recorded at the beginning of the tests and immediately after the stress changes meant that the values of ϵ_u, ϵ_L and ϵ_i are difficult to measure accurately. However, in the present work, there was no discernible difference between ϵ_i, ϵ_u and ϵ_L for the same stress change (Fig. 9). The present results thus indicate that the deformation processes con-

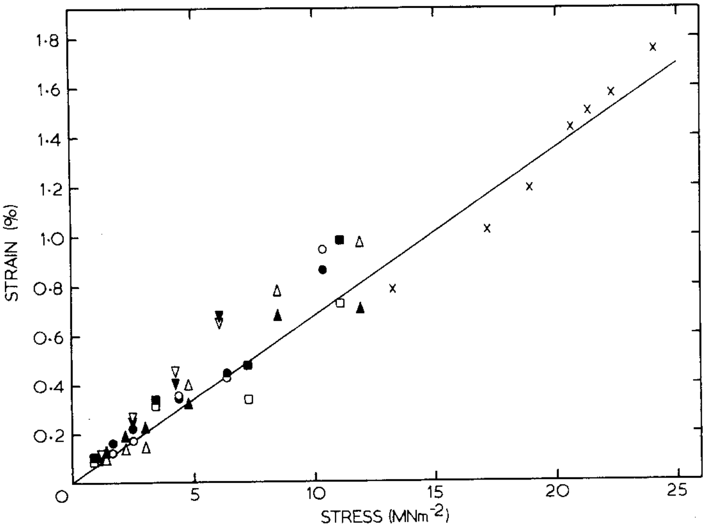

Figure 9 The variation of the instantaneous strain with stress for polypropylene at 293 K. The stress dependence of the initial specimen extension on loading (X) is shown in relation to the values obtained when the applied stress is changed suddenly by a small amount (σ) during steady-state creep at 24.13 MNm^{-2} (▾▿), 22.41 MNm^{-2} (▴▵), 21.38 MNm^{-2} (▪□) and 20.69 MNm^{-2} (●○). The instantaneous strains, ϵ_u, recorded on reducing the stress by various amounts during creep are presented as open symbols while the extensions, ϵ_L, observed on increasing the stress by different amounts are shown as closed symbols.

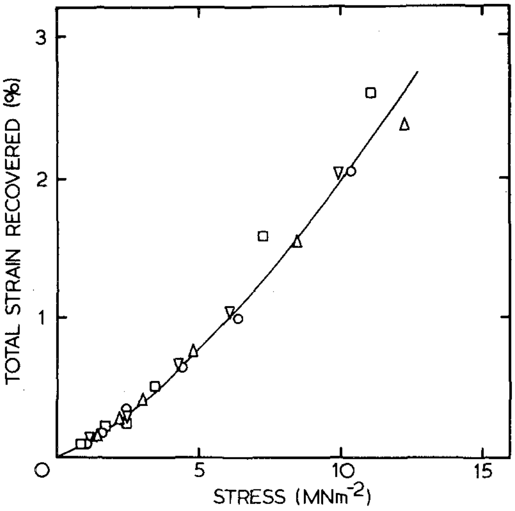

Figure 10 The dependence of the total strain recovered, ϵ_R, on the magnitude of the stress reduction made during steady state of creep of polypropylene at 24.13 MNm^{-2} (▽), 22.41 MNm^{-2} (△), 21.38 MNm^{-2} (□) and 20.69 MNm^{-2} (○).

trolling the instantaneous changes in specimen length are the same for both loading and unloading with polypropylene.

The occurrence of a period of negative creep immediately after the instantaneous specimen contraction, ϵ_u, on reducing the stress has usually been explained on the basis that creep occurs under an effective stress, σ_{eff}, as [18]

$$\sigma_{eff} = (\sigma - \sigma_{int}), \qquad (10)$$

where σ_{int} is the internal stress developed during creep. With this approach, the strain–time characteristics recorded following a load reduction would be expected to depend on the magnitude of the remaining stress, σ_R. When $\sigma_R > \sigma_{int}$, the effective stress would be positive so that a new, slower, positive creep rate should be obtained immediately after the instantaneous specimen contraction, ϵ_u. When $\sigma_R = \sigma_{int}$, the effective stress would be zero giving a zero creep rate. Only when σ_{eff} becomes negative (when $\sigma_R < \sigma_{int}$) should a period of negative creep be observed.

The behaviour observed after stress reductions for polypropylene does not, however, appear to be compatible with a model based on creep being controlled by the effective stress, σ_{eff}. The existence of a period of negative creep means that the specimen deforms as if suddenly subjected to a compressive force. No direct conclusions can be drawn from measurements of the creep rates before and after the stress decrease since the creep behaviour of thermoplastics differs in tension and compression [6, 19]. Yet, when the total recovered strain, ϵ_R, was defined as the difference between the strain recorded immediately prior to the stress reduction and the value observed when the creep rate became zero under the reduced load, σ_R, the relationship between ϵ_R and $\Delta\sigma$ for polypropylene tested at different values of the applied stress appears to extrapolate to zero (Fig. 10). The elastic stresses developed in the materials during secondary creep must therefore be equal to the full applied stress, so that periods of negative creep are obtained (Fig. 8) even with very small stress reductions ($\Delta\sigma \simeq 0.05\sigma$). As negative creep continues the elastic stresses are gradually reduced to the value appropriate to the reduced stress level. Eventually the positive creep rate attained becomes equal to the secondary rate expected for an uninterrupted test carried out entirely at the lower stress level (Fig. 11), showing that full recovery of the creep properties takes place for the materials studied in the present work.

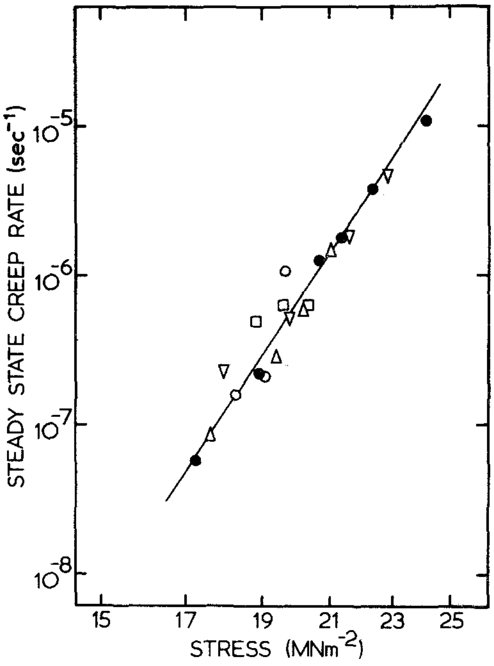

Figure 11 The stress dependence of the constant creep rate eventually established following various stress reductions during steady-state creep of polypropylene at 24.13 MNm^{-2} (▽), 22.41 MNm^{-2} (△), 21.38 MNm^{-2} (□) and 20.69 MNm^{-2} (○). For comparison, the dependence of the steady state creep rate, $\dot{\epsilon}_s$, on applied stress, σ, for uninterrupted tests is presented as closed symbols.

5. Conclusions

The strain–time behaviour observed during primary and secondary creep of high density polyethylene and polypropylene at 293 K can be adequately described by the equation

$$\epsilon = \epsilon_0 + \epsilon_p [1 - \exp(-K\dot{\epsilon}_s t)] + \dot{\epsilon}_s t.$$

However, during the initial 10 to 20% of the primary creep life, deviations occur when the creep rate decreases more rapidly than is predicted by this equation.

The stress dependence of the secondary creep rate and also the strain rate sensitivity of the flow stress can be expressed as

$$\dot{\epsilon}_s \alpha \sigma^n,$$

with $n \simeq 16$ for polypropylene and $n \simeq 11$ for polyethylene. Moreover, the relationship shown to exist between the primary creep parameters and the secondary creep rates over a wide range of strain rates demonstrates that the same deformation processes control the creep behaviour throughout primary and secondary creep.

Reduction of the applied stress during secondary creep to continue the test at a reduced stress level, σ_R, is invariably accompanied by a rapid specimen contraction, followed by a period of negative creep before a new positive creep rate is eventually established. Since periods of negative creep were recorded even after very small stress reductions ($\sigma_R \simeq 0.96\sigma$), the elastic stresses developed in the material during secondary creep are equal to the full applied stress, σ, so that the material behaves as if suddenly subjected to a net compressive stress immediately following stress reductions. These elastic stresses decrease gradually following the load reduction until they become equal to the reduced stress level, σ_R, at which stage the positive creep rate established is identical to the secondary creep rate recorded in an uninterrupted test carried out entirely under a stress equal to σ_R. Full recovery of the creep properties is therefore attained after stress reductions in the range $\sigma_R \simeq 0.96$ to 0.54σ carried out during secondary creep.

Acknowledgements

The author is indebted to the Science Research Council for a maintenance grant during the period of this research, and also to Dr. B. Wilshire for discussions concerning the manuscript.

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Received 17 December 1979 and accepted 8 July 1980