A comparison of tensile, compressive and torsional creep in isotropic and oriented polyethylene

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An account is given of the creep behaviour, under tensile, compressive and torsional loading conditions, of oriented samples of a linear polyethylene (LPE) which have been produced by the process of hydrostatic extrusion. A comparison is made between the behaviour of oriented and isotropic LPE material and, with biomedical applications in mind, between the performance of the linear LPE polyethylene and the ultra-high molecular weight polyethylene grade commonly used in prostheses.

1. Introduction

The research reported in this paper forms part of an ongoing study of the properties and structure of oriented polymers produced by solid phase deformation processes such as hydrostatic extrusion and tensile drawing. Because of the possible application of such materials in long-term load bearing situations, it is clearly valuable to establish some understanding of their creep and recovery behaviour. During the last few years very extensive studies of the tensile creep behaviour of oriented polyethylenes have been undertaken, with particular regard to the effects of molecular weight, draw ratio, copolymer content and more recently electron irradiation [1–4]. The results have provided guidelines for the use of such materials in civil engineering applications [5, 6].

The present research is concerned with the creep behaviour of oriented linear polyethylene rods in tensile, compressive and torsional loading. Because of the thickness of these rods it was possible for compression samples to be obtained having their axes transverse to the principal direction of orientation (i.e. the extrusion direction) thereby enabling the creep behaviour in this transverse direction to be studied. Tensile and torsion tests could only be performed on samples having their orientation axes parallel to the loading axis. Most of the tests were conducted at 37° C, (nominal body temperature) because the motivation for the research had been provided by possible application of these stiffer and stronger polyethylenes as components of protheses within the human body.

2. Experimental procedure

2.1. Preparation of materials

The oriented polyethylene rods were produced by hydrostatic extrusion and full details of the production procedures are given in previous publications [7, 8].

The polymer selected was Rigidex 006-60 ($\bar{M}_w$ = 135 000; $\bar{M}_n$ = 25 500) a polyethylene homopolymer manufactured by BP Chemicals Ltd, and supplied in pellet form. An isotropic billet was produced by melt extrusion at about 200° C into a vertical cylinder. A weighted piston situated over the molten extruded polymer allowed uniform filling of the mould, which was transferred to an oven at 110° C, and subsequently cooled slowly to room temperature. In this way completely void-free billets were prepared.

The isotropic billets were machined to a suitable diameter for producing final products at extrusion ratios (λ) of 5 and 10, with final product diameters of either 15.5 or 23.8 mm. The hydrostatic extrusion was performed at 100° C, with a conical die of semi-angle 15°. The pressure transmitting fluid was castor oil, which was heated to the required temperature in the extrusion vessel and pressurized by a ram intensifier, backed by a hydraulic pump.

In addition to isotropic and oriented Rigidex 006-60, it was also considered appropriate, for comparison purposes, to examine the behaviour of the ultra-high molecular weight grade of polyethylene (UHMWPE), RCH 1000, produced by Ruhr-Chemie AG, which has been used successfully in bioengineering applications since 1960. This was supplied in the form of compression moulded blocks by High Density Plastics Ltd, Todmorden, UK.

2.2. Apparatus

Uniaxial tensile and compressive creep tests were performed by exploiting the lever and cage system of apparatus originally designed at the laboratories of ICI [9]. A modified version of the dead-loading creep apparatus was constructed at the authors' laboratory by Wilson *et al.* [10].

The main load is applied to the specimen by means of a lever arm pivoted so as to give a load magnification

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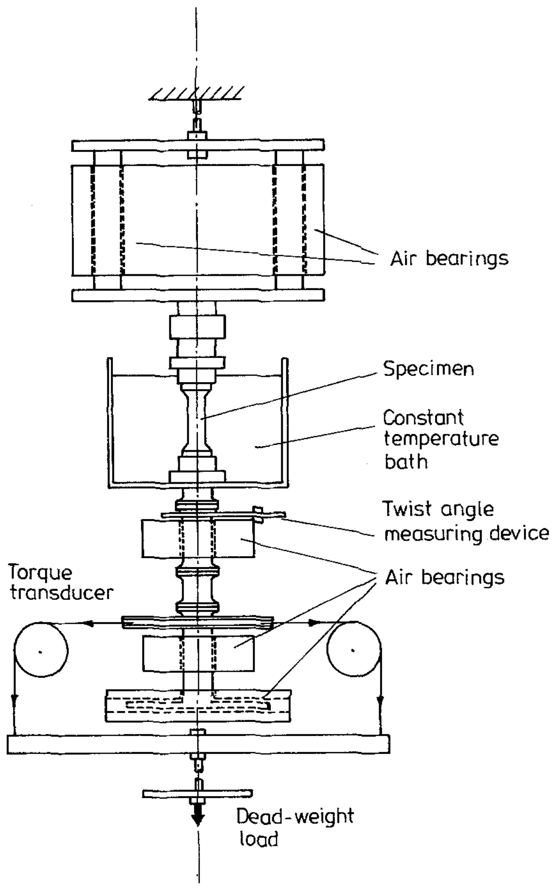

Figure 1 Schematic diagram of torsional creep apparatus.

of 12 : 1 in the loading cage. The movement of part of the cage in relation to a fixed stage, allows for compressive and tensile tests to be conducted relative to the fixed stage. For compressive tests, flat mild steel platens, with their surface perpendicular to the direction of loading, are used. For tensile tests, the specimen is held in pin chucks.

An insulating chamber is lowered over the loading cage to provide a temperature-controlled environment. A heater and fan built into the roof of the chamber, provide the required temperature with air circulation between an inner and outer chamber.

In the torsion tests, the dead-loading creep apparatus employed was originally designed and used by Smith [11]. This apparatus is shown diagrammatically in Fig. 1. The specimen is held within a perspex tank of water which is maintained at a constant temperature using a temperature controlled recirculation system. The dead-weight loading is transmitted by means of phosphor-bronze tapes and pulleys to be converted into a couple applied about the main vertical axis of the apparatus. The resulting torque is transmitted via the torque transducer to the lower specimen grip. The upper specimen grip is supported in a set of air bearings which allows frictionless axial movement but prevents axial rotation. Air bearings are also employed to support the lower part of the torque transmitting and measuring assembly. These bearings provide both vertical and sideways frictionless support. In order to eliminate vertical axial loading on the specimen a system of counterbalancing is used to offset the weights of the various components of the apparatus.

The principal variables to be measured during the torsion test are the applied torque and the resulting angle of twist as functions of time. The torque transducer based on two pairs of electrical resistance rosette gauges provides the means for measuring the torque and the angle of twist is also measured electrically by a potentiometer which uses a nylon gear mechanism to amplify the rotation of the lower end of the specimen.

2.3. Creep measurements

For axial compressive creep studies, cylindrical specimens of diameter 10 mm and height 40 mm were machined from the initial materials. They were placed axially between the smooth flat platens of the compression cage of the apparatus. For the study of compression perpendicular to the direction of extrusion, for which the height of the machined sample was restricted, two cylinders each of diameter 9.5 mm and height 19 mm were machined and stacked axially. This arrangement, which maintained a 4 : 1 aspect ratio, was shown to behave identically with a single cylinder in tests on an isotropic sample. It was considered that the single polymer/polymer interface in the stack of two cylinders, being equidistant from the platens, would not cause a strain discontinuity at this interface.

For the study of tensile creep, cylinders of length 120 mm and diameter 4 mm were machined. They were gripped in pin chucks to as to give a uniformly loaded length of 100 mm. Deformation at the ends could be neglected since strain was measured from the central 10 mm section of the specimens. In both tension and compression an Instron strain gauge extensometer was used to measure strain. This had an accuracy corresponding to $\pm 10^{-3}$ mm in 10 mm, i.e. 0.01% strain [12].

For torsion testing, the specimen was cylindrical of diameter 9 mm and gauge length 40 mm. Beyond the gauge length, the specimen diameter was increased to 16 mm tapering to 24 mm. The specimen was gripped along the tapered section ensuring that deformation was restricted to the gauge length section. The accuracy of the measurement of the angle of twist is ± 0.01 rad, which is equivalent to a strain of $\pm 0.1\%$. While this accuracy is somewhat less than that for the tensile and compressive measurements, it will be noted that the minimum tensile and compressive creep strains are for material having draw ratio $\lambda = 10$ and are $\sim 0.1\%$, whereas in torsion the lowest strains measured are much greater and are $\sim 1\%$.

2.4. Pretreatment of test specimens

The machined polymer test specimens would have experienced a complex pattern of mechanical deformation as a result of the various forming processes undergone during manufacture. Some residual stresses would be expected to exist within the specimens. They were therefore first annealed at a temperature of 60° C for a period of 50 h in deionized water before testing. They were then allowed to cool slowly to room temperature. The process of relaxation of residual stresses would have thus been accelerated. However, being

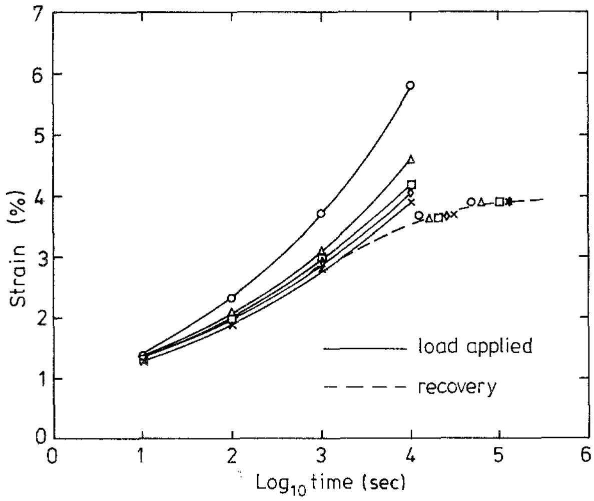

Figure 2 Influence of conditioning – variation of strain with time for five successive loading cycles for extruded R006-60 at applied stress (σ = 13.3 MPa). Load number 1 2 3 4 5
Symbol 0 △ □ ◊ ×

well below the melting temperature, physical changes in the structure are avoided.

Following the annealing procedure, the specimen was mounted in the loading apparatus and allowed to acclimatize at the test temperature for a period of several hours. The specimen was then conditioned; a procedure first considered necessary by Leaderman [13] and subsequently found to be satisfactory for oriented polymers, including oriented polyethylenes, in research investigations [1] for many years.

The specimen was loaded at the highest stress that was to be applied, and for the maximum period of creep testing. This maximum stress was always chosen to be below the yield plot for the material. The period of creep was invariably 10^4 sec. The specimen was then allowed to recover for a period of 10^5 sec. This procedure of creep and recovery was repeated, usually four times, until eventually, the creep behaviour was reproducible. Interestingly, the recovery profile following the first loading was little different from that following the final loading even though the creep strain may have decreased by 50%.

When repeatability was achieved, it was found that the "conditioned" specimen could then be subjected to any stress for any time provided that the maximum strain reached at the end of the conditioning programme, was never exceeded. The change in creep and recovery during conditioning is illustrated in Fig. 2 in all modes of deformation. The magnitude of unrecovered strain during conditioning decreased as a consequence of orientation of the polymer. This augurs well for the dimensional stability of components produced from the oriented material.

3. Results

3.1. Creep and recovery in compression

From the viewpoint of possible biomedical applications, the creep and recovery behaviour in compression is of greatest interest. In Figs. 3a and b the compressive behaviour of the isotropic polymers RCH 1000 and R006-60 are compared. At the maximum stress levels comparatively large strains of 3 to 5% were observed for creep times of 10^4 sec, but the recovery 10^5 sec after removal of the load is seen to be virtually complete. The ultra-high molecular weight RCH 1000 is approximately twice as compliant as R006-60. This difference can be attributed to its lower crystallinity, and as shown in Fig. 3b the results for the two polymers can be brought into coincidence by simply scaling the magnitude of the response for the R006-60 by a constant factor of two.

The oriented samples show a large reduction in the magnitude of the creep response (Figs. 4a and b). This result could have been anticipated for compressive creep in the axial or longitudinal direction, because it is well known that the stiffness is increased in this direction by orientation. It is particularly interesting to note that the creep response is also substantially reduced for compression transverse to the orientation direction. The significance of this result will be discussed later.

In recent discussions of the creep behaviour of oriented polyolefines, with regard to their engineering applications [1–5], it has been proposed that it is useful to examine the creep data in terms of log strain rate–strain plots. Sherby and Dorn [14] showed that this representation is appropriate for the case of isotropic polymethylmethacrylate, where the creep response

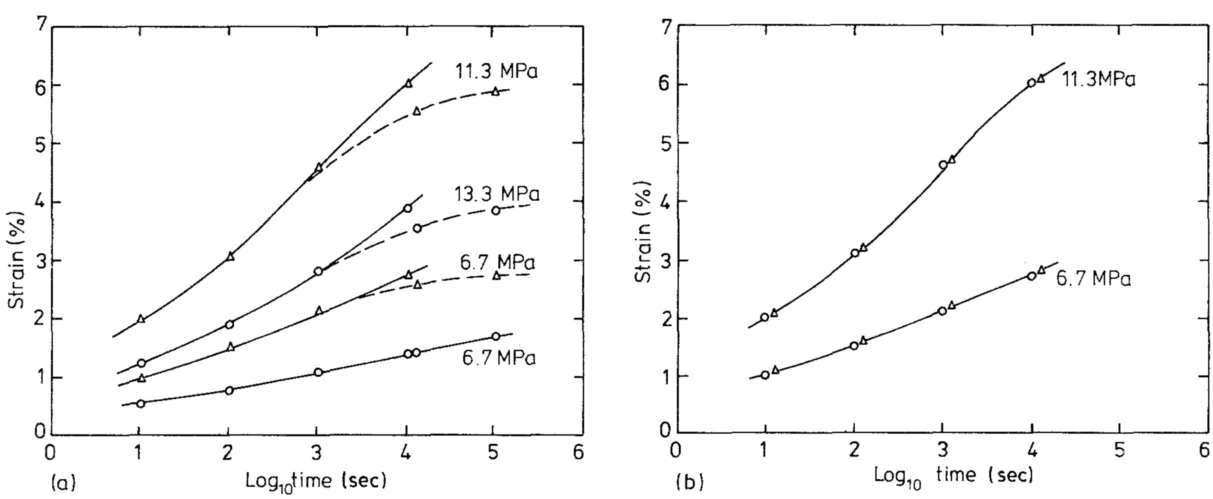

Figure 3 Compressive creep of two isotropic polyethylenes. (a) Creep and recovery at different stress levels: △ = RCH 1000; ○ = R006-60; and (b) scaling factor applied to creep results: △ = RCH 1000 (× 1.0); ○ = R006-60 (× 2.0).

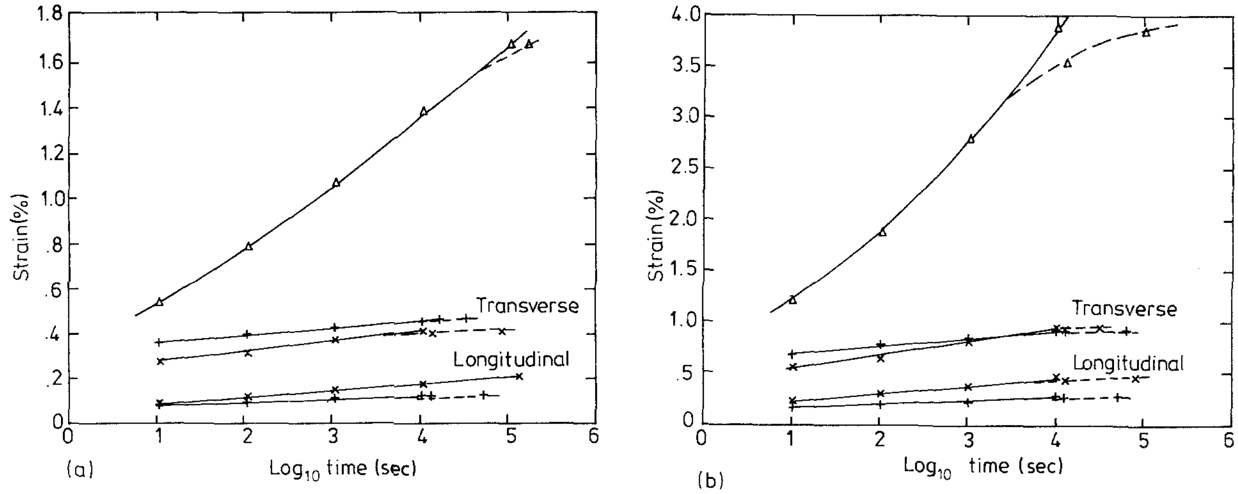

Figure 4 Compressive creep of extruded R006-60 as a function of compressive stress and orientation direction. △, isotropic ($\lambda = 1$); ×, $\lambda = 5$; +, $\lambda = 10$. (a) Applied stress = 6.7 MPa and (b) applied stress = 13.3 MPa.

approximates to a thermally activated process. Wilding and Ward [1] showed that, although the log strain rate–strain plots for oriented polyethylenes were not consistent with this very simple model, the creep rates at high stresses reached a limiting constant value, and that these plateau creep rates were consistent with a simple thermally activated process. Moreover, from a practical engineering viewpoint, it can be considered that there is a critical stress level below which plateau creep behaviour is not observed. For stresses below this critical stress the creep rate falls continuously, so that the creep strain reaches a limiting constant value and failure does not occur. If the load is removed the recovery for such stress levels is virtually complete.

In Fig. 5 Sherby–Dorn plots are shown for the isotropic polymers and for the extruded materials at a stress level of 11.3 MPa. These plots show the distinctions between the different materials very clearly, the advantages of the extruded materials being very evident.

3.2. Creep and recovery in tension

The creep and recovery behaviour of the isotropic polymers in tension is very similar to their behaviour in compression, and as can be seen from Figs. 6a and

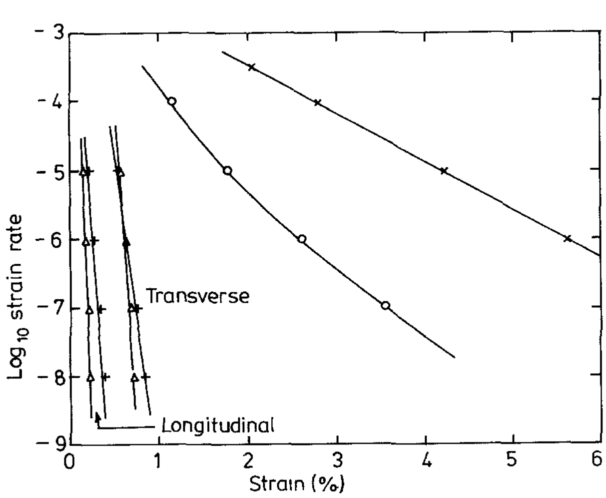

Figure 5 Strain rate behaviour in compression (Sherby–Dorn plots) for RCH 1000 (×, $\lambda = 1$) and extruded R006-60 (○, $\lambda = 1$; +, $\lambda = 5$; △, $\lambda = 10$) at 11.3 MPa applied stress.

b the results can be very nearly brought together by scaling the responses at 10 sec by a factor of two. The effect of orientation is again to reduce the magnitude of the response but as can be seen by comparing Figs. 7a and b with Figs. 4a and b, which shows the compression results, there are some differences in detail. In particular, for a draw ratio of 5 the creep response is larger in tension than compression, whereas the reverse is true for a draw ratio of 10.

In view of the extensive studies of the creep behaviour of drawn LPE fibres, it appeared to be of some value to establish the links with drawn monofilaments and again to present the present data in terms of Sherby–Dorn plots. In Figs. 8 and 9a and b, respectively, Sherby–Dorn plots are shown for oriented monofilaments and the extrudate of R006-60 at $\lambda = 10$, and for the isotropic polymers and the R006-60 extrudates. It can be seen that the filaments show a plateau creep response for the stress level of 100 MPa, and that the stress levels appropriate for biomedical applications are well below the critical stress, the extrudate data showing excellent continuity with those for monofilaments. It is of some interest to note that the RCH 1000 appears to be approaching the plateau creep behaviour in tension at a stress level of 11.2 MPa. The oriented extrudates show a very marked reduction in tensile creep, similar to that observed in compression.

3.3. Creep and recovery in torsion

In the study of torsion, the maximum shear stress τ_R was calculated from the torque–twist curves, using the Nadai [15] analysis; the maximum shear strain taken as $\gamma = r\theta/l$, where θ is the angle of twist and r and l are the radius and length of the solid cylinder, respectively. Typical isochronous torque–twist curves, for the isotropic R006-60, together with the corresponding calculated stress–strain curves are shown in Fig. 10. From such calculated isochronous stress–strain curves, the creep strain could be calculated as a function of time for a given stress level. Results for the isotropic polymers are shown in Fig. 11 and are very similar to those found for tension and compression, with a similar factor of two between the responses for the two materials.

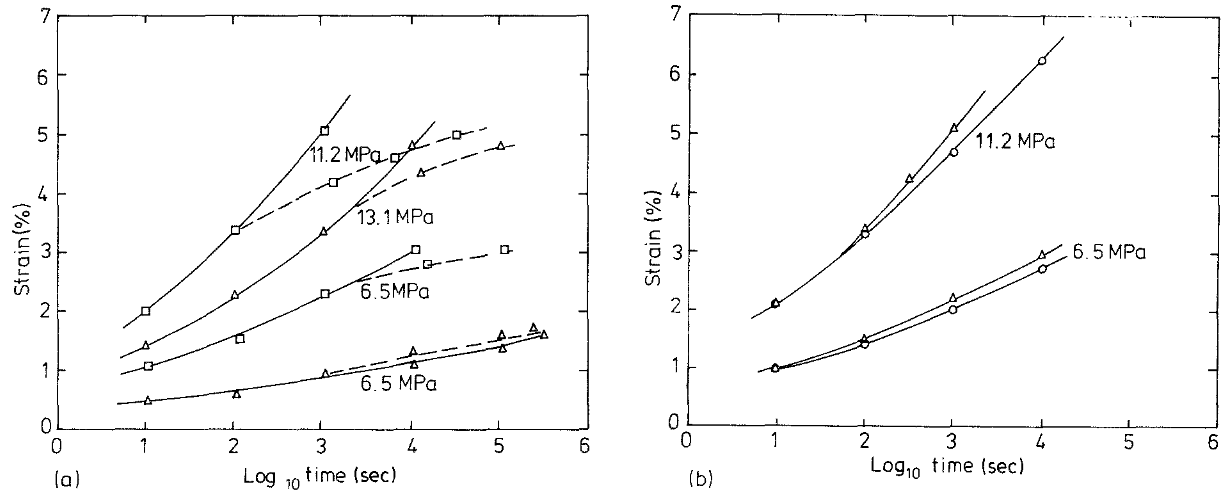

Figure 6 Tensile creep of two isotropic polyethylenes. (a) Creep and recovery at different stress levels: □ = RCH 1000; △ = R006-60; and (b) scaling factor applied to creep results: △ = RCH 1000 (× 1.0); ○ = R006-60 (× 2).

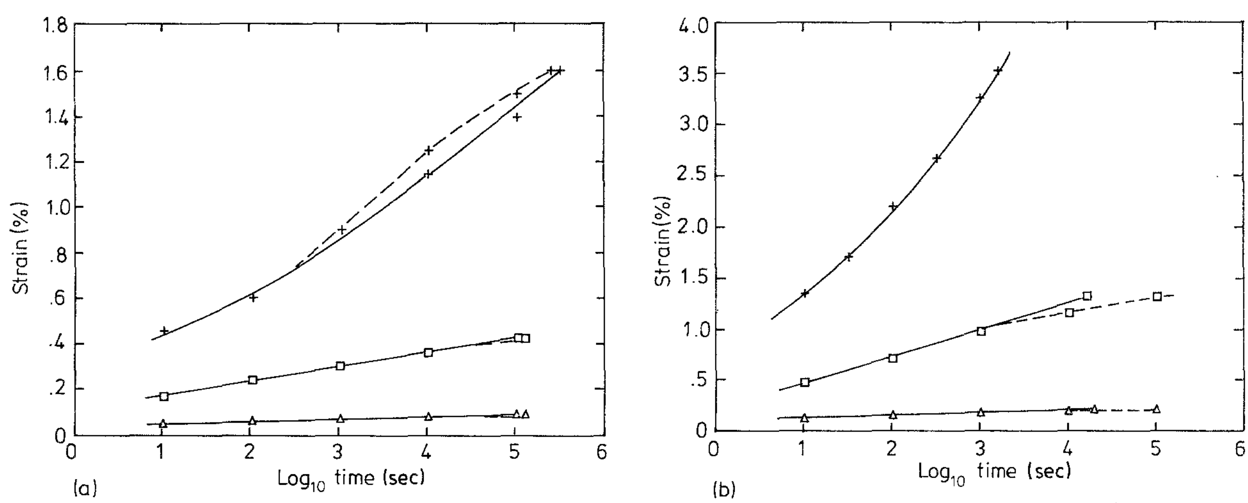

Figure 7 Tensile creep of extruded R006-60 as a function of tensile stress and extrusion ratio. +, isotropic (λ = 1); □, λ = 5; △, λ = 10. (a) Applied stress = 6.5 MPa and (b) applied stress = 13.1 MPa.

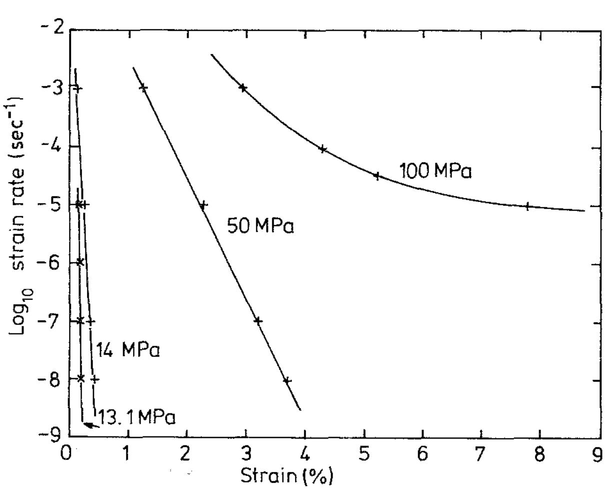

Figure 8 Strain rate behaviour in tension — comparison between filaments (+) and rods (×) of R006-60 produced by tensile drawing and hydrostatic extrusion, respectively, at reduction ratios of λ = 10.

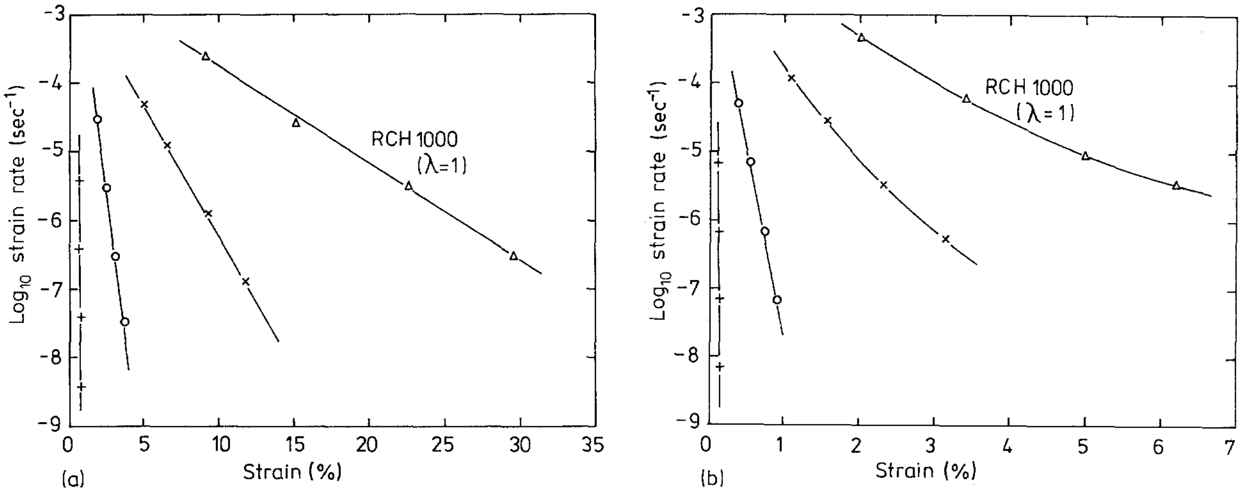

Figure 9 Strain rate behaviour in tension for RCH 1000 (△, $\lambda = 1$) and extruded R006-60 (×, $\lambda = 1$; ○, $\lambda = 5$; +, $\lambda = 10$) at two different stress levels. (a) Applied stress = 6.5 MPa and (b) applied stress = 11.2 MPa.

The torsional creep data for the R006-60 extrudates are shown in Figs. 12a and b. These results show little or no difference in torsional creep between the isotropic material and a draw ratio of 5, but a marked reduction in response for a draw ratio of 10 extrudate. These differences between materials are also seen very clearly in the Sherby–Dorn plots (Fig. 13).

4. Discussion

4.1. Isotropic polymers

It has been proposed that the creep behaviour of isotropic polymers is essentially determined by the deviatoric component of the applied stress, with the hydrostatic component playing a minor role, analogous to its influence on yield behaviour [16].

Benham and Mallon [17] have been prominent in attempting to relate shear and uniaxial creep behaviour in polymers using the concept of an equivalent strain proportional to the octahedral shear strain, γ_{oct} and an equivalent stress proportional to the octahedral shear stress, τ_{oct}. To take into account the difference between behaviour in tension and compression the mean value of the octahedral shear strains in tension and compression was equated to the octahedral strain in pure shear.

For tension and compression

$$\tau_{oct} = (2^{1/2}/3)\sigma$$

and for torsion

$$\tau_{oct} = (2/3)^{1/2}\tau$$

where σ is the applied tensile or compressive stress, and τ is the applied shear stress. For tension and compression $\tau_{oct} = [2(2^{1/2})/3]\varepsilon(1 + \nu)$ where ε is the axial strain and ν is the Poisson's ratio.

The simplest assumption is to take $\nu = 0.5$ (constant volume deformation) in which case

$$\gamma_{oct} = (2)^{1/2}\varepsilon$$

It is also plausible to assume that $\nu = 0.4$ for the initial elastic strain ε_1 but takes the value 0.5 during the subsequent creep deformation, which produces a strain ε_2. We then have

$$\gamma_{oct} = \frac{2(2)^{1/2}}{3}(1.4\varepsilon_1 + 1.5\varepsilon_2)$$

For torsion, $\gamma_{oct} = (2/3)^{1/2}\gamma$ where γ is the shear strain (taken as the maximum shear strain at the surface of the cylinder).

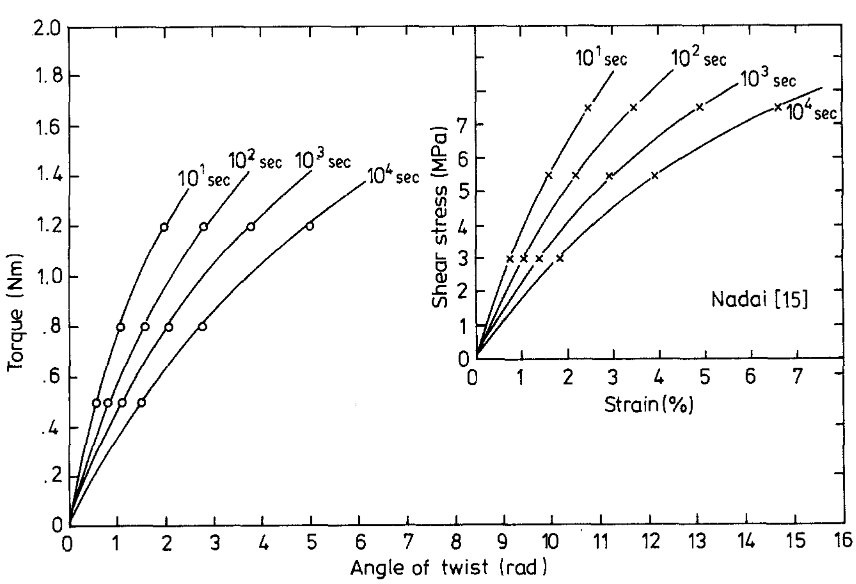

Figure 10 Isochronous torque–twist and shear stress–shear strain curves for isotropic R006-60.

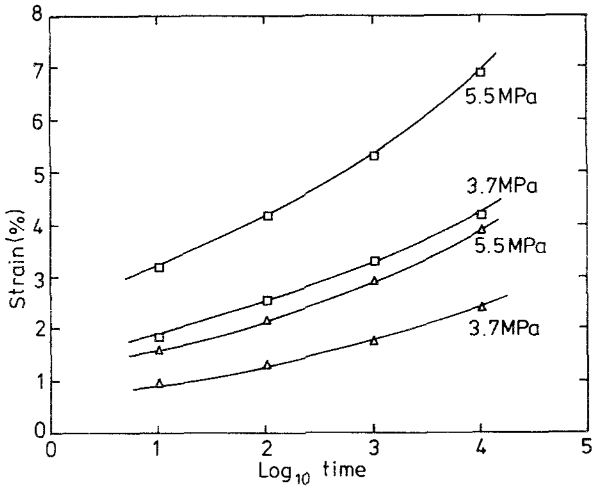

Figure 11 Torsional creep of two isotropic polyethylenes at different stress levels: □ = RCH 1000; △ = R006-60.

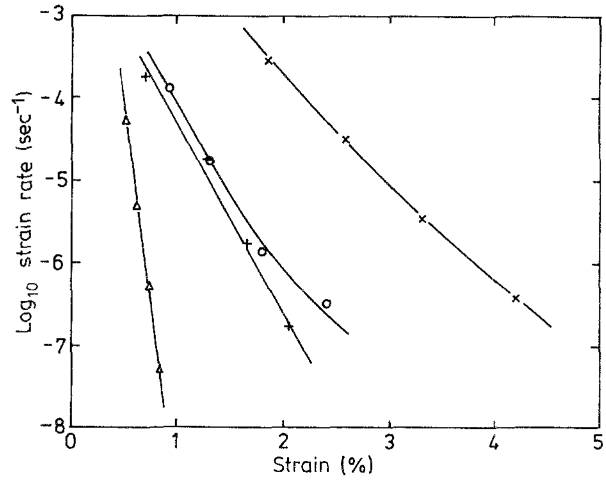

Figure 13 Strain rate behaviour in torsion for RCH 1000 (λ = 1) and extruded R006-60 (○, λ = 1; +, λ = 5; △, λ = 10) at an applied shear stress of 3.7 MPa.

Results are presented in Fig. 14 for the isotropic polymers in terms of the octahedral shear strain as a function of time, combining the data already presented for tension and compression with interpolated data for torsion, using the procedures described above. It can be seen that there is good support for the proposition that the octahedral shear stress is the key parameter in determining the creep behaviour of isotropic polyethylene. This result, taken together with the effectiveness of the scaling procedure to bring the data for the two different polymers into coincidence, suggests that shear of the amorphous regions controls the creep response of the isotropic polymer in the temperature range studied. It is interesting to speculate that this is interlamellar shear, as discussed with regard to the relaxation processes of linear polyethylene.

From previous work it might have been anticipated that the influence of the hydrostatic component of stress would mean that the creep strains in tension were greater than those observed in compression, with those observed in torsion being intermediate. The present results can hardly be claimed to support this trend, and in fact the differences between the three stress fields is less than $\pm 2\%$ for R006-60 and less than $\pm 7\%$ for RCH 1000.

4.2. Oriented extrudates

It is apparent from the initial survey of results presented above that for all the modes of deformation examined, tensile, compressive and torsion, there is a marked reduction in the creep response for the oriented extrudates compared with the isotropic polymers.

For the anisotropic polymers, previous studies indicate that it is likely that the tensile, compressive and shear modes of stress will activate several different mechanisms of deformation by different amounts. Consequently, the simple approach of equivalence of strain on the octahedral shear plane for an equivalent octahedral shear stress is no longer applicable. It is, however, interesting to make the empirical comparison between deformation in the three modes as in terms of the octahedral shear strain and shear stress. Results for R006-60, λ = 5 and 10, shown in Figs. 15a and b, respectively, indicate that the shear mode of deformation is a predominant mode of deformation in the oriented samples. In the λ = 5 sample the response is very comparable to that of the isotropic R006-60 polymer. There is some reduction of shear response in the λ = 10 sample, as would be expected from dynamic mechanical studies of comparable materials.

An interesting comparison can be made between

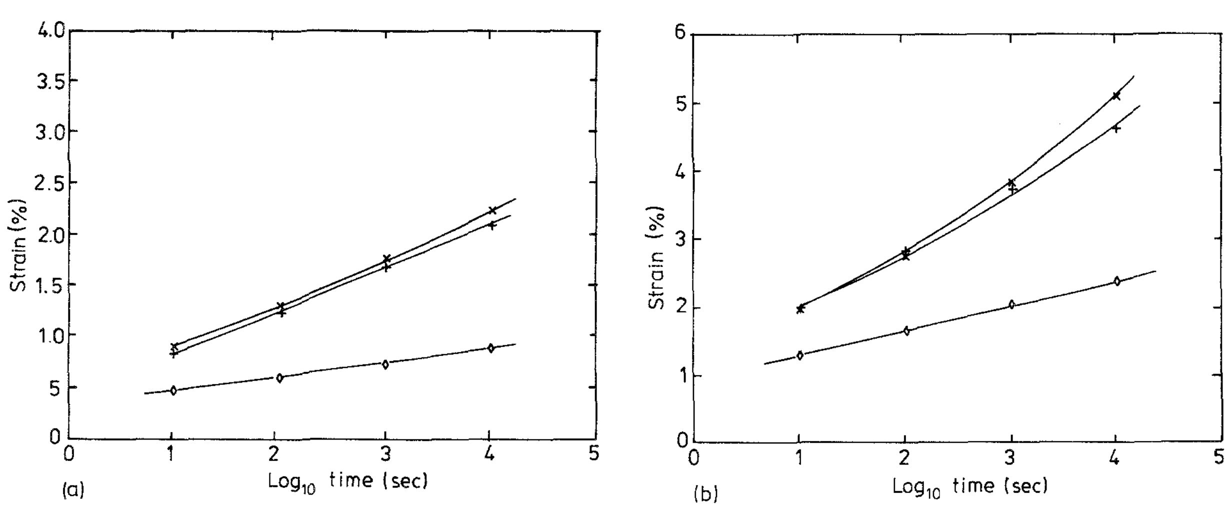

Figure 12 Torsional creep of extruded R006-60 as a function of shear stress and extrusion ratio: ×, λ = 1; +, λ = 5; ◊, λ = 10. (a) Applied stress = 3.7 MPa and (b) applied stress = 6.5 MPa.

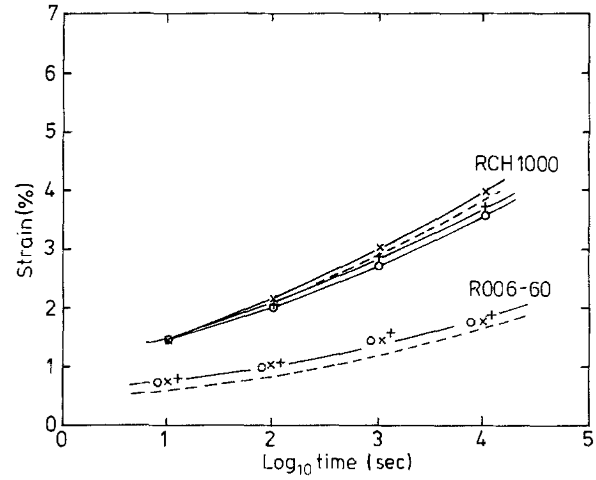

Figure 14 Octahedral shear strain as a function of time derived from tensile, (×), compressive (+) and torsional (○) creep results, for isotropic RCH 1000 and R006-60. Octahedral shear stress = 3.0 MPa, i.e. σ = 6.5 MPa or τ = 3.7 MPa. ——— ν = 0.5; – – – ν = 0.4 to 0.5.

tension and compression parallel to the extrusion direction. In the $\lambda = 5$ sample the tensile mode shows a greater response than the compressive mode. As already noted the creep behaviour in the shear mode for this sample is very close to that of the isotropic polymer. These results are in agreement with the conclusion of Duckett [18] that polymers drawn to low and intermediate draw ratios show similar yield behaviour to isotropic polymers i.e. the differences between tension and compression are dominated by the effect of the hydrostatic component of stress rather than the molecular orientation.

For the $\lambda = 10$ sample, the situation is reversed, and whereas the deformation in the compressive mode is similar to that for the $\lambda = 5$ sample, the deformation in the tensile mode is much reduced. This is analogous to the yield behaviour of oriented polymers. It is easier for oriented polymers to yield in compression by buckling and shear modes of deformation than in tension where there is intensive strain-hardening, because the stress is acting on an already extended structure.

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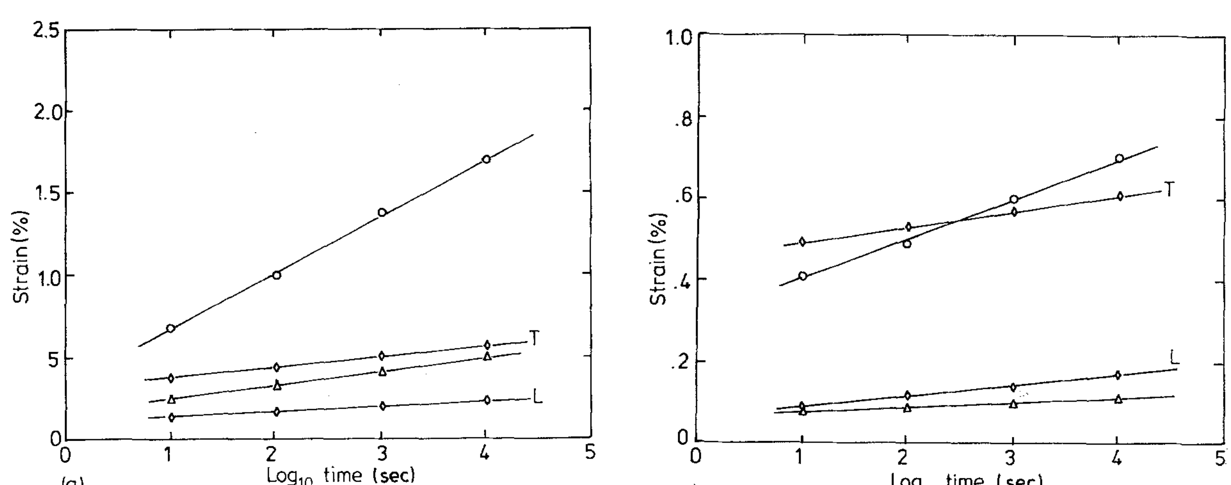

Figure 15 Octahedral shear strain as a function of time derived from tensile (△), compressive (◊, T = transverse; L = longitudinal) and torsional (○) creep results for extruded R006-60. Octahedral shear stress = 3.0 MPa, i.e. σ = 6.5 MPa or τ = 3.7 MPa. (a) Extrusion ratio $\lambda = 5$ and (b) extrusion ratio $\lambda = 10$.