Uniaxial tensile creep behaviour of ultra high molecular weight linear polyethylene

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The present paper examines the room temperature constant-load uniaxial tensile creep response of two ultra high molecular weight linear polyethylene (UHMW LPE) materials and compares it with that of a normal molecular weight linear polyethylene (NMW LPE). It was found that at all stress levels examined, the magnitude of creep deformation is significantly higher in UHMW LPE than in NMW LPE. Possible reasons for this behaviour are explored. Potential techniques for improving the tensile creep behaviour (i.e. decreasing the creep deformation) of LPE are discussed.

INTRODUCTION

Ultra high molecular weight linear polyethylene (UHMW LPE) is reported to have a unique combination of excellent properties[1-3]*. It is known for the two properties (a) outstanding wear and abrasion resistance[8-11] and (b) impact resistance[3,6]; fatigue behaviour[1,12] and chemical resistance[4] are also emphasized. It was shown to possess an excellent impact–fatigue behaviour[13-15]. UHMW LPE is believed to be an ideal material for numerous different applications such as gears, bushings, liners for chutes, bins and conveyors, wear plates, prostheses, etc.[16-21]. An extended discussion on the influence of molecular weight and of molecular weight distribution on various mechanical properties of polyethylene and several other polymers is available in a review article[22].

The high molecular weight that imparts outstanding properties to LPE also means that UHMW LPE cannot be processed conventionally because of the extremely high melt viscosities. However, the fabrication of UHMW LPE will not be discussed here. Interested readers can find information on this topic elsewhere[23-29]. The present paper deals with the 1000-h *constant-load* uniaxial tensile creep response of two UHMW LPE materials and compares it with that of a normal molecular weight linear polyethylene (NMW LPE). Although, various aspects of the tensile creep response of polyethylenes have been reported in the literature[30-43], to the author's knowledge, not much work has been published on the tensile creep behaviour of UHMW LPE. Limited data are presented in reference 7 which discusses the tensile, compressive and flexural creep behaviour of UHMW LPE as a function of temperature. The present paper examines the uniaxial tensile creep behaviour at 23°C of UHMW LPE and NMW LPE as a function of applied stress.

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* Although the definition is quite arbitrary and there is no universal agreement in the scant literature on what constitutes ultra high molecular weight[4-6], UHMW LPE in the present work would imply LPE with a weight-average molecular weight in excess of 1.5 million. It might be added that most of the reported values of molecular weight in the ultra high molecular weight range are only approximate because of the problems associated with measuring extremely high molecular weights for LPE[3,7].

MATERIALS

The two UHMW LPE and one NMW LPE materials examined, together with some of their physical and thermal properties are listed in *Table* 1.

EXPERIMENTAL PROCEDURES

All specimens were machined from 3.2 mm thick sheets compression-moulded according to the following procedure. The platens of a Pasadena Hydraulics Press were preheated to 200°C. The picture-frame mould was placed between two Mylar(R) films with back-up chrome plates. (The mould-cavity contained a preweighed quantity of polymer powder.) The entire assembly was maintained between the press platens for 3 min with a low nominal pressure (< 50 psi*) applied to the moulding. Keeping the temperature at 200°C, the nominal pressure on the moulding was then raised to ~500 psi*, and the new conditions were maintained for a further 3 min. The press platens were then watercooled to ~50°C in approximately 10 min. While cooling the platens, the nominal pressure was gradually raised to and maintained at ~1000 psi to minimize 'sink marks'. The pressure was then released in ~45 s and the moulding recovered.

Constant-load 1000-h uniaxial tensile creep response of these materials was measured according to the procedures outlined in ASTM Standard D-674. The 3.2 mm thick specimens were milled to 1.25 cm width. Elongations on a 5 cm gauge-length were measured with a cathetometer having an accuracy of ~ ±0.0005 cm. Five different stress levels ranging from 750 to 1700 psi were employed. For each material, a single (fresh) specimen was tested at each different load (or nominal stress) level. Five specimens of each material were used in the present work.

Uniaxial tensile load-elongation behaviour of all three materials was examined according to ASTM procedure D-638.

In an attempt to destroy the past thermal history, all test specimens in the present work were pre-conditioned for 48 h at the final testing conditions (23°C and 50% relative humidity).

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* 1 psi $= 6.895 \times 10^3$ N/m^2

Table 1 Materials employed, together with some of their physical and thermal properties

Material	Reported intrinsic viscosity (dl/g)	Density (g/cm^3) Powder	Moulded sheet	Differential scanning calorimetry (d.s.c.) peak melting temperature ($^\circ$C)	Scanning (d.s.c.) Heat of fusion (cal/g)	Crystallinity calculated from density (%)	heat of fusion (%)	Young's modulus ($\times 10^{-5}$ psi)
UHMW LPE-A	19.8[a]	0.962	0.928	134	32.6	48.6	46.8	1.0
UHMW LPE-B	14.8[b]	0.962	0.933	135.5	33.3	52.1	47.8	1.2
NME LPE	2.7[c]	0.962	0.962	136.5	49.0	72.9	70.3	1.7

[a] Although the exact molecular weight is difficult to measure, both materials have an $M_W > 2 \times 10^6$, as measured by light scattering
[b] $M_W \sim 207\,000$, as measured by g.p.c. *MI* ~ 0.3. M_W = weight-average molecular weight. *MI* = melt index
[c] All materials supplied by the Dow Chemical Company; the UHMW LPE resins are experimental materials
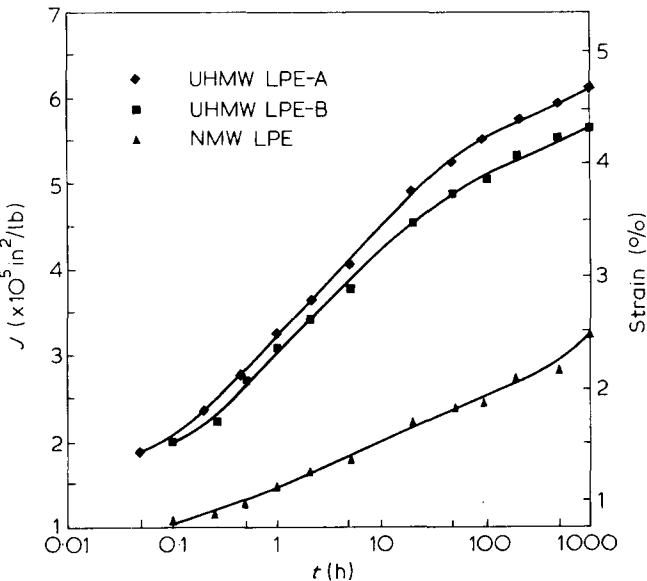

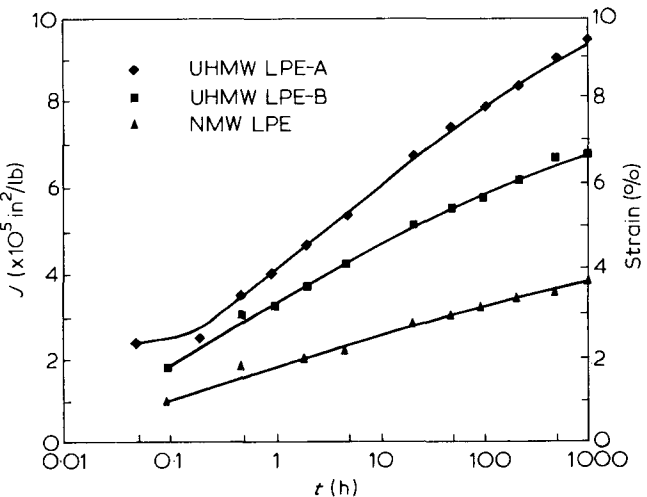*Figure 1* Uniaxial tensile creep compliance and strain *versus* time for all three materials; σ_0 = 750 psi; 1 psi = 6.895 x 10^3 N/m^2

Figure 2 Uniaxial tensile creep compliance and strain *versus* time for all three materials; σ_0 = 1000 psi; 1 psi = 6.895 x 10^3 N/m^2

Figure 3 Uniaxial tensile creep compliance and strain *versus* time for all three materials; σ_0 = 1200 psi; 1 psi = 6.895 x 10^3 N/m^2

EXPERIMENTAL RESULTS

Creep compliance data

The experimental results are presented in *Figures* 1–5 in the form of creep compliance *versus* time plots. For quick and easy reference, the percent creep strains experience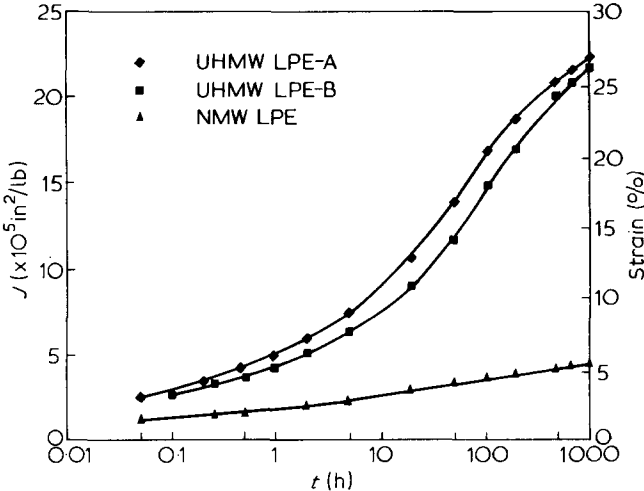d by the samples are also indicated on these figures. Each figure shows the creep response of all three materials evaluated at a given nominal stress level 750–1700 psi. The tensile creep compliance, $J(t)$, at time t is defined as:

$$J(t) = \varepsilon(t)/\sigma_0 \qquad (1)^*$$

where, $\varepsilon(t)$ = total nominal or engineering strain at time t,

$$\sigma_0 = \text{(nominal) applied stress}$$

$$= \frac{\text{Applied load}}{\text{Original cross-sectional area}}$$

The results are intentionally presented in the form of $J(t)$ *versus* log t curves to offset the effect of slight differences in the actual stress levels in the specimens.

Figures 1–5 show that the J *versus* log t curves for UHMW LPE, at least for the lower stress levels examined, tend to assume a sigmoidal shape. Thus, as t or log t increases, the slope of these curves increases for some time followed by a decrease. This final decrease in slope may be indicative of the presence of a 'network structure' which is quite conceivable in an UHMW LPE due to the extensive chain entanglements present in such a polymer.

* As is apparent from the definition, J is calculated on the basis of *total* strain which includes the instantaneous or load-on strain (time-independent component) and the creep strain (time-dependent component).
Also, the results reported here are for constant-load and not cosntant-stress creep; no attempt has been made to apply any correction for the change in cross-sectional area of the specimen with time.
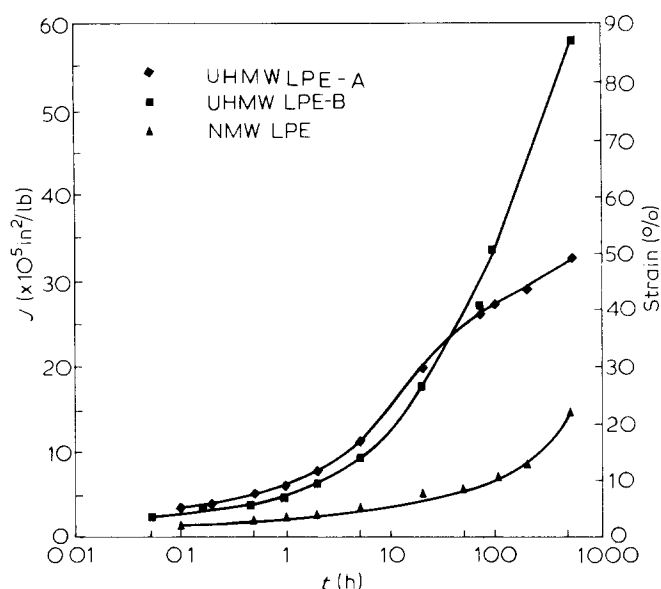
Figure 4 Uniaxial tensile creep compliance and strain *versus* time for all three materials; σ_0 = 1500 psi; 1 psi = 6.895 x 10^3 N/m^2; test purposely discontinued after 500 h
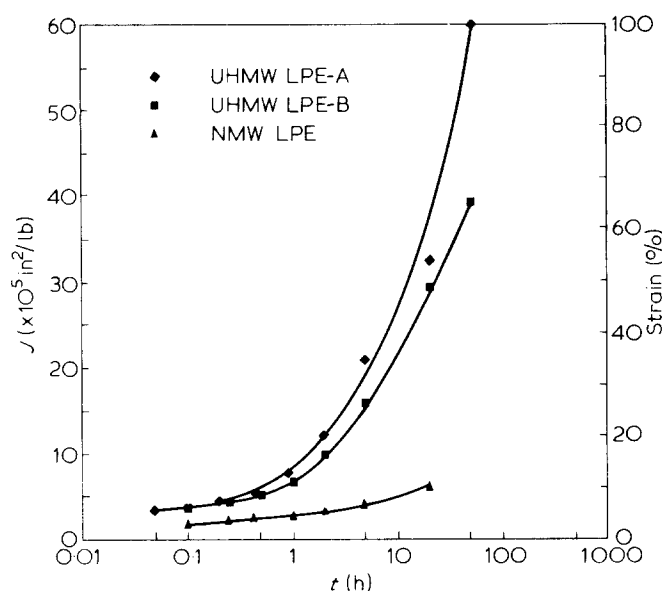

Figure 5 Uniaxial tensile creep compliance and strain *versus* time for all three materials; σ_0 = 1700 psi; 1 psi = 6.895 x 10^3 N/m^2; test purposely discontinued after 50 h

It can be seen from *Figures* 1–5 that at all stress levels examined, both UHMW LPE materials undergo much higher uniaxial tensile creep deformation at a given time than does the NMW LPE. The creep deformation at any given time and stress level is directly related to the molecular weight. For example, UHMW LPE-A, which has the highest molecular weight, has the highest creep deformation whereas NMW LPE, which has the lowest molecular weight has the lowest creep deformation. The higher creep deformation in UHMW LPE results from its lower degree of crystallinity.

The data of *Figures* 1–5 are replotted in *Figures* 6 and 7. *Figure* 6 shows for UHMW LPE-A the log–log plot of *J* *versus* *t* with stress-level as a parameter. The identical plot for the other UHMW LPE had qualitative features very similar to those for UHME LPE-A and is not shown

here. The similar plot for NIW LPE is shown in *Figure* 7. As seen from these figures, *J* *versus* *t* curves are not independent of the applied stress-level. Thus, neither NMW LPE nor UHMW LPE exhibit linear viscoelastic behaviour. For both these materials, the creep deformation becomes increasingly non-linear in stress, as the stress level is increased.

Deformation behaviour of specimens during creep testing

All the specimens were visually observed during and after creep testing. A set of samples of different materials tested at 1700 psi is shown in *Figure* 8. At 1700 psi, all the specimens except the NMW LPE specimen underwent localized necking followed by neck propagation. Oblique shear bands, although not discernible in *Figure* 8, could be observed in the necked region on the surface of these samples. At 1500 psi, all materials except UHMW LPE-A underwent localized necking. The NMW LPE sample (the only one of this material to experience necking phenomenon) also exhibited the stress-whitening commonly observed in tension. The specimens at lower stress levels deformed uniformly and did not undergo the inhomogeneous deformation of the necking phenomenon.

Equations describing creep behaviour of LPE

Several empirical equations have been proposed to describe the creep response of polymeric materials[44,45]. These equations take one of the following two general forms:
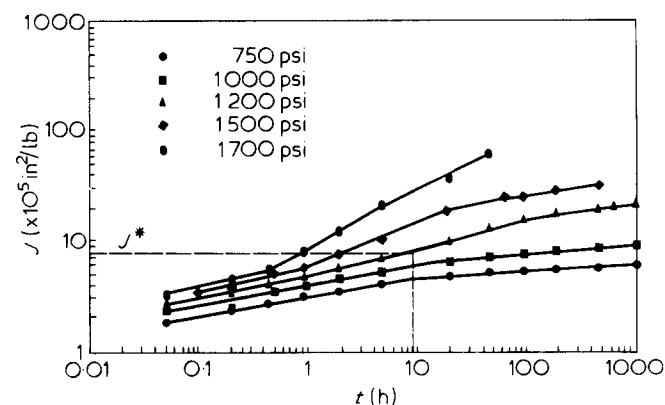

Figure 6 Log–log plot showing for UHMW LPE-A the uniaxial tensile creep compliance *versus* time at different stress levels
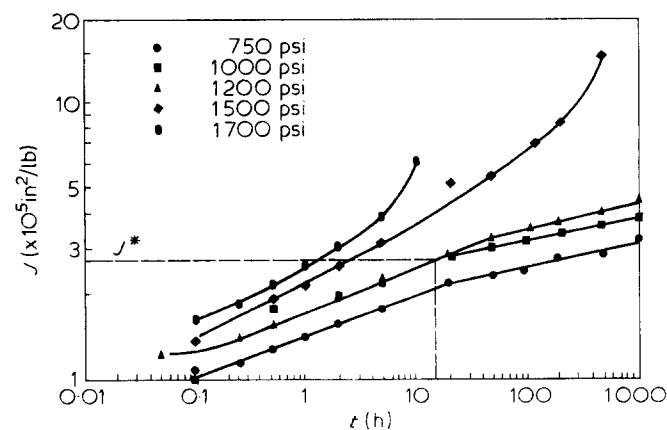

Figure 7 Log–log plot showing for NMW LPE, the uniaxial tensile creep compliance *versus* time at different stress levels
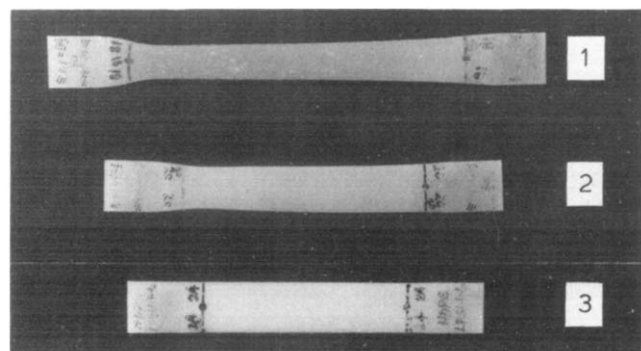
Figure 8 Photograph showing specimens tested in uniaxial tensile creep at a nominal stress of 1700 psi: 1. UHMW LPE-A; 2. UHMW LPE-B; 3. NMW LPE
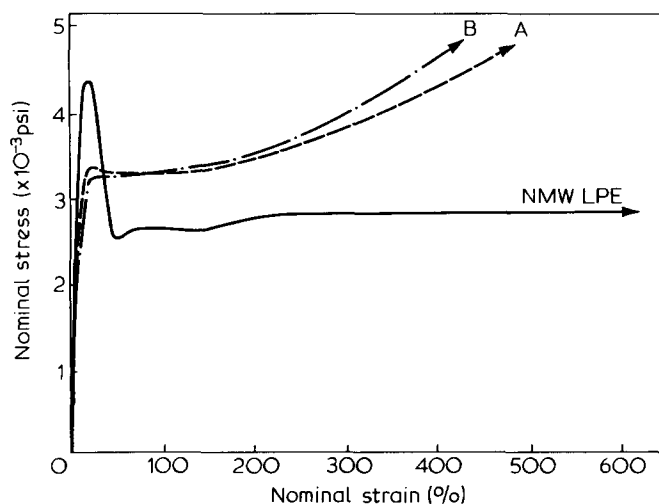

Figure 9 Uniaxial tensile nominal stress—strain curves for all three materials; 1 psi = 6.895 x 10^3 N/m²

$$\varepsilon = F(\sigma, t) \qquad (2)$$

or

$$\varepsilon = f_1(\sigma) \cdot f_2(t) \qquad (3)$$

Where F, f_1 and f_2 are functions of the variables indicated above.

An examination of *Figures* 6 and 7 reveals that at low compliances $(J \leqslant J^*)$, the stress-dependence and time-dependence of creep deformation are essentially separable, as indicated by almost parallel curves on the log J versus log t plots[†] with stress-level as a parameter; i.e. at low compliances $(J \leqslant J^*)$, the creep response for both UHMW LPE (except at the highest stress level) and NMW LPE can be described by an equation of type (3) above. At higher compliances $(J > J^*)$, the dependence of creep strain on stress and time is inseparable. Also, the creep compliance–time relationship appears to be linear on log–log plots (*Figures* 6 and 7), for approximately two decades of time, at least on the lower stress levels. In other words, the creep response of both UHMW LPE and NMW LPE in this time region can be represented by an equation of the form:

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† In view of equation (1), the log ε vs log t curve would simply be displaced from the log J vs log t curve by an amoung log σ_0 along the ordinate.

$$\varepsilon = f_1(\sigma) \cdot t^\kappa \qquad (4)$$

where $\kappa = d \log \varepsilon/d \log t$ is the slope of log strain–log time curves of *Figures* 6 and 7. It appears that for the three lower stress levels, the creep strain–time relationship even at higher strain levels can be described by equation (4) with a different value of κ. This type of equation was employed by Buchdahl and Nielsen[46] to describe the creep behaviour of PVC blends. These authors observed that at least when the stress-level is not too high, the value of κ is independent of the stress-level. That seems to be the case with the UHMW LPE (*Figure* 6) and the NMW LPE (*Figure* 7) at the three lower stress levels.

Tensile stress–strain behaviour

Uniaxial tensile stress–strain behaviour of all three materials was also evaluated and the results are presented in *Figures* 9 and 10, respectively, in the form of nominal stress–strain curves and a corresponding set of tested samples.

As seen from these figures, the UHMW LPE materials have lower yield strengths but much higher ultimate strength than the NMW LPE. In the post-yield region, while the NMW LPE undergoes 'perfectly plastic' response with cold drawing, the UHMW LPE exhibits strain-hardening. The NMW LPE, in going from the unoriented isotropic state to the post-yield oriented anisotropic state does so in a non-uniform manner. It deforms by the initiation and propagation of a localized macroscopic neck with concomitant stress-whitening. The UHMW LPE, however, does not exhibit any localized macroscopic necking or the stress-whitening. In addition, immediately after fracture, the UHMW LPE specimens relax and undergo considerable twisting and kinking, *Figure* 10. That the latter behaviour results from non-homogeneous deformation and recovery in UHMW LPE specimens is quite likely, especially in view of the numerous macrodefects, which appear to be partially-fused 'gels', observed in the UHMW LPE mouldings. These defects assume an elongated shape in the deformed state.

DISCUSSION

The data presented clearly show that at all stress levels examined, the creep deformation at a given time is much higher in UHMW LPE than in NMW LPE. The two counteracting factors that could explain this effect and that immediately come to one's mind are: (a) molecular weight, and (b) overall level of crystallinity and any
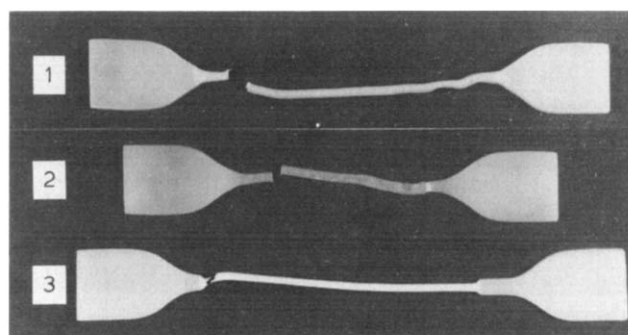

Figure 10 Photograph showing specimens tested in uniaxial tension: 1. UHMW LPE-A; 2. UHMW LPE-B; 3. NMW LPE
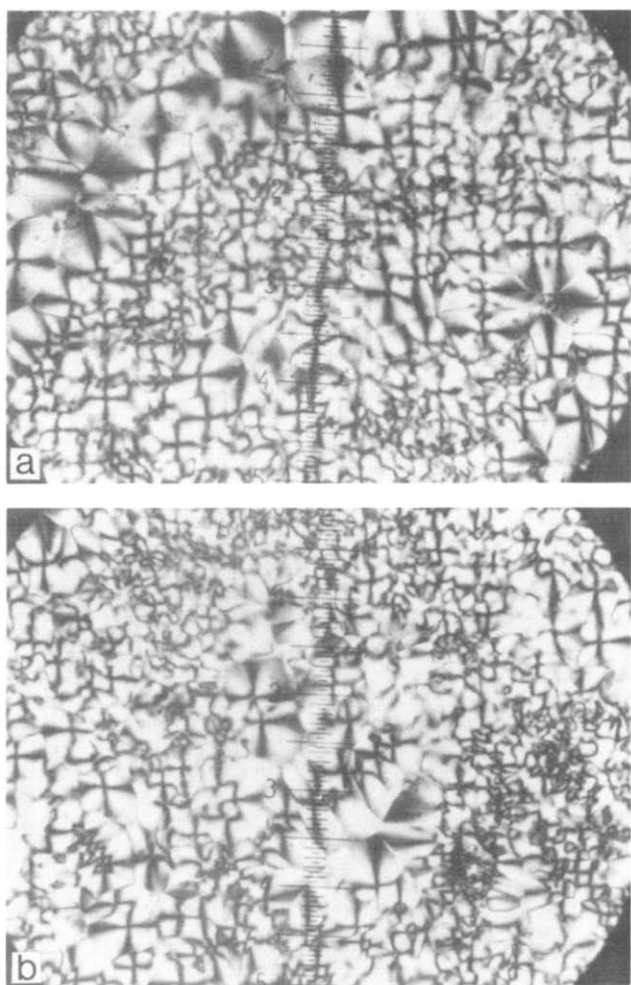
Figure 11 Optical photomicrographs showing the morphological features at two different regions of a sample of UHME LPE-A; (1 smallest scale division = 2 μm)

properties. The tensile moduli for all three materials are given in *Table 1*, while the yield stresses are shown in *Figure 9*. UHMW LPE-A can be seen to have the lowest tensile modulus, while the NMW LPE has the highest modulus. UHMW LPE has a lower tensile yield stress than does NMW LPE. The lower tensile modulus and yield strength of UHMW LPE can be expected to induce higher creep strains in this material.

Quantitatively isolating the individual effects of molecular weight and crystallinity on tensile creep would require additional creep measurements on samples having identical levels of crystallinity but differing only in molecular weight, and/or on samples with the same molecular weight but different crystallinity. These measurements could not be included in the present study due to the unavailability of either of the above two kinds of samples.

In view of the data presented, UHMW LPE will not be a suitable candidate for applications where creep is a primary requirement. In order to improve the creep resistance of UHMW LPE, one could conceivably adopt one or a combination of different techniques mentioned below:

(a) Introduce an optimum number of cross-links in the system; one possible means of achieving this could be through exposure to high energy radiation.
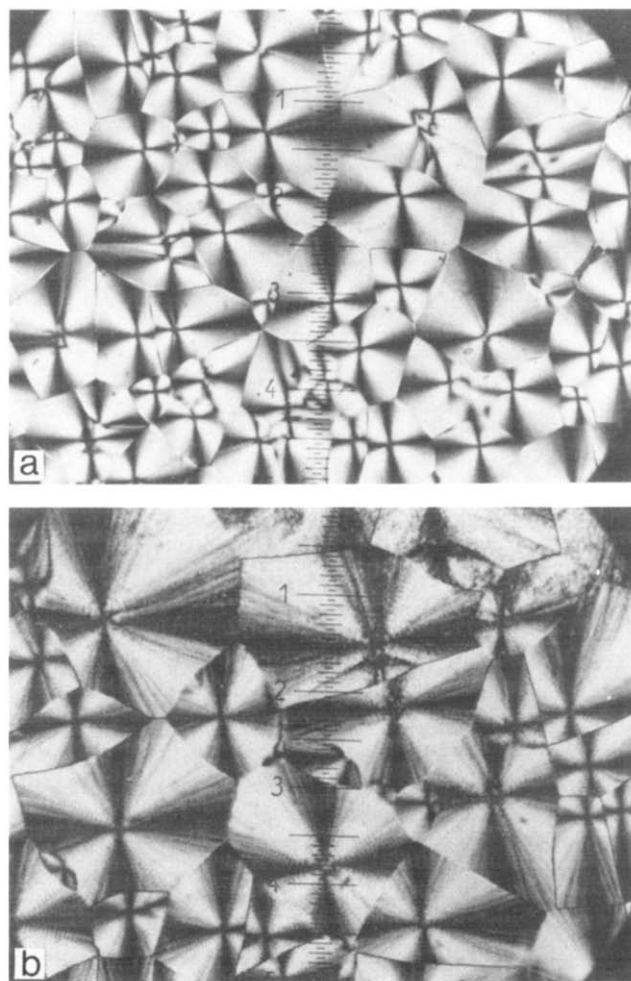
possible difference in the crystalline morphology. It would seem that the increased chain entanglements due to high molecular weight would tend to retard and reduce creep deformation in UHMW LPE. However, in view of the larger creep deformation at any time *t* in UHMW LPE, the lower levels of crystallinity that have been observed in UHMW LPE (*Table* 1) and also any possible differences in the crystalline morphology seem to be the overriding factors. Crystalline content has been observed in the past to influence the creep behaviour of other polyethylenes. For example, the creep resistance was reported in the past to improve with increasing density for low density polyethylene[33,36], ethylene-propylene copolymer and NMW LPE[36]. In the present case, besides the differences in the degree of crystallinity, some differences in the crystalline morphology have also been observed. Thin films of UHMW LPE-A and NMW LPE were examined in an optical microscope between crossed polaroids and the results are shown in *Figures* 11 and 12, respectively. The NMW LPE shows well-defined spherulitic structure while in UHMW LPE, the spherulites are not as well-defined. Some of the spherulites that can be recognized in UHMW LPE are much smaller. Apart from the presence of some spherulites, the UHMW LPE sample examined also shows some small unresolved morphological features over large portions of the sample. Interrelated with the degree and type of crystallinity are the mechanical

Figure 12 Optical photomicrographs showing the spherulitic structure at two different regions of a NMW LPE; (1 smallest scale division = 2 μm)

(b) Increase the level of crystallinity; the crystallinity of the compression-moulded sheets examined in this study is quite low.

(c) Use fillers and reinforcements.

In addition to improving the creep response of UHMW LPE, these techniques will also enhance several other properties such as heat distortion, stiffness and strength, etc. The important consideration would, of course, be if the creep property enhancement can be realized without adversely affecting other properties such as impact response and wear and abrasion resistance.

The tensile creep response of radiation-exposed UHMW LPE has in fact been examined and the findings will be published in a forthcoming paper[47].

CONCLUSIONS

At all stress levels examined, the magnitude of the uniaxial tensile creep deformation ($t \leqslant 1000$ h) in UHMW LPE is significantly higher than in NMW LPE. UHMW LPE is not a suitable material for applications where creep resistance is a primary requirement. The higher creep deformation in UHMW LPE results from its lower level of crystallinity. *Quantitatively* isolating the individual effects of molecular weight and crystallinity on the tensile creep response would require additional measurements. At the low stress levels investigated, the stress-dependence and time-dependence of creep deformation are essentially separable both in UHMW LPE and NMW LPE.

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