Energy criterion for modelling creep rupture of high-density polyethylene

B. W. Cherry and Teoh Swee Hin*
Department of Materials Engineering, Monash University, Clayton, Victoria 3168, *Australia*
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A technique involving the use of a three-element mechanical model with a critical stored energy criterion modelled accurately the creep rupture time of two types of high-density polyethylene (HDPE) specimens. The upper stress limit where the specimen ruptured immediately on application of load and the lower stress limit where the specimen sustained the load indefinitely were also features of the model. These two limits were found to depend on the resilience, elastic modulus and anelastic modulus of the HDPE.

Keywords Activated rate process; anelasticity; creep rupture; model; polyethylene; Reiner–Weissenberg criterion

INTRODUCTION

In general the methods of polymer creep rupture prediction can be divided into three groups. The first group depends upon the basic assumption that rupture is an activated rate process. In this approach the log(time to fracture) is related to the applied stress by structural parameters such as activation volume and activation energy[1]. Extensive reviews have been made on this group of methods[2,3]. The second group consists of empirical methods based on experimental observation, curve fitting and extrapolation techniques[4,5]. The third group is based on the continuum mechanical theories which assume that the material under study is homogeneous and isotropic. Here it is further assumed that there is a specific material property such as uniaxial tensile strength, total elongation, or energy storage capability that determines the failure of the stressed specimen. Often to aid visualization of the solutions of the differential equations which are used to describe the viscoelastic response (elastic, anelastic and plastic), mechanical models consisting of a combination of springs and dashpots are used[6].

Recently the failure characteristics of thermoplastics have been related successfully to an energetic limit. Bruller[7] used the Reiner–Weissenberg thermodynamic theory of strength[8] to predict the transition from linear to non-linear viscoelasticity. For the limit of crazing and fracture, a modified version of the above theory was used.

In the majority of the techniques used in creep rupture prediction, difficulty is encountered in predicting:

(1) the upper stress limit where the specimen ruptures immediately on application of load, and

(2) the lower stress limit where, although the specimen creeps, it sustains the load indefinitely without rupture.

Both these limits are extremely important for technological reasons. Current work on creep rupture prediction of high-density polyethylene showed that a technique involving the use of a mechanical model in conjunction with the Reiner–Weissenberg energy failure criterion modelled accurately the rupture time as well as the above two limits.

THE PROPOSED MODEL

The purpose of mechanical modelling is to provide an easy visualization of the solutions of the differential equations which are used to describe the elastic, anelastic and plastic responses to an applied stress. The definitions of these responses can be confusing. For example, the instantaneous strain recorded in a constant-load experiment has been described as due to the elastic response[9]. However, Polakowski and Ripling[10] suggested that the initial instantaneous strain consists of both an elastic and a plastic component. Fotheringham and Cherry[11] have shown that for a high-density polyethylene the permanent strain starts immediately when an external stress is applied. Mindel and Brown[12], bearing in mind the confusion associated with these terms, prefer to define arbitrarily the elastic, anelastic and plastic strain, since their definitions depend on the speed of measurement. In this paper, the elastic response is defined as the instantaneous response that is completely recoverable on stress removal and the word 'instantaneous' is arbitrarily taken to mean 'occurring within 1 s or less'. The assumption made in this definition is that, during the time interval 1 s after removing the load, other responses are small compared to the elastic response. The anelastic response can be defined as the non-instantaneous (time-dependent) recovery process (sometimes referred to as delayed elasticity[6]) after the stress is removed from a stressed specimen. This definition was also used by Lloyd and McElroy[13] and Mindel and Brown[12]. Plastic re-

* Now at Department of Mechanical and Production Engineering, National University of Singapore, Kent Ridge, Singapore 0511

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sponse can be defined as the permanent or residual response, which does not change significantly with time, left in a specimen after the deforming stress is removed.

Fotheringham and Cherry[14] have shown that the deformation behaviour of high-density polyethylene can be described by a three-element model which consists of a storage element in series with a parallel arrangement of a dissipative element and another storage element. A three-element model of this kind allows one to partition the applied stress between the recovery stress, which is responsible for bringing about the recovery process, and the effective stress, which is responsible for plastic flow. Some further evidence for the validity of such a partition has been reported by Shinozaki and Sargent[15].

In this paper a simple three-element model which consists of a Hookean spring in series with a parallel arrangement of another Hookean spring and an Eyring dashpot (see *Figure 1*) is used. The definition of an Eyring dashpot has already been described by a number of authors[14,16,17]. This dashpot can be considered to be filled with a non-Newtonian liquid whose viscosity is stress dependent according to a hyperbolic sine function. The proposed model can be used to predict creep rupture if one assumes that rupture occurs when the total stored energy in the elastic and anelastic spring reaches a critical value. This is equivalent to the Reiner–Weissenberg energy criterion of failure[8]. Referring to *Figure 1*, the above concept can be described mathematically as:

$$\int_0^{\varepsilon_a^*} \sigma_{re}\, d\varepsilon_a + \int_0^{\varepsilon_e^*} \sigma_{ap}\, d\varepsilon_e = R \tag{1}$$

where ε_a^* = anelastic strain at rupture, ε_e^* = elastic strain at rupture, σ_{re} = recovery stress, σ_{ap} = applied stress and R = resilience of the material. The word 'resilience' is defined as the material property related to the intrinsic free energy which can be stored elastically in the volume element of the material.

Using $\sigma_{re} = \varepsilon_a E_a$ where ε_a and E_a are the anelastic strain and anelastic modulus respectively, the first integral in equation (1) can be evaluated as $\varepsilon_a^{*2} E_a/2$. Under creep conditions, the applied stress is constant. Hence, by using $\sigma_{ap} = \varepsilon_e^* E_e$ where E_e is the elastic modulus, the second integral in equation (1) can be evaluated as σ_{ap}^2/E_e. Thus equation (1) becomes after rearrangement:

$$\varepsilon_a^* = \left[\frac{2}{E_a}\left(R - \frac{\sigma_{ap}^2}{E_e}\right)\right]^{1/2} \tag{2}$$

It has been shown by Fotheringham and Cherry[14] that the anelastic strain rate ($\dot{\varepsilon}_a$) can be expressed in terms of the effective stress (σ_{ef}) as:

$$\dot{\varepsilon}_a = K \sinh(\beta\sigma_{ef}) \tag{3}$$

where K is a function of the activation energy and β is a function of the activation volume (V) and is equal to $V/2kT$ (k and T represent Boltzmann's constant and then absolute temperature, respectively). The anelastic strain rate can also be expressed as:

$$\dot{\varepsilon}_a = \dot{\sigma}_{re}/E_a \tag{4}$$

The recovery stress rate ($\dot{\sigma}_{re}$) can be related to the applied ($\dot{\sigma}_{ap}$) and effective stress rate ($\dot{\sigma}_{ef}$) as:

$$\dot{\sigma}_{re} = \dot{\sigma}_{ap} - \dot{\sigma}_{ef} \tag{5}$$

Hence, assuming that $\dot{\sigma}_{ap} = 0$ (creep experiment) and using equations (3)–(5), the following expression can be derived:

$$\int_{\sigma_{ap}}^{\sigma_{ef}} \frac{d\sigma_{ef}}{\sinh(\beta\sigma_{ef})} = -\int_0^t E_a K\, dt \tag{6}$$

where t = time. The limits in equation (6) derive from the fact that in the three-element model shown in *Figure 1* it can be seen that at zero time the applied stress is acting only on the Eyring dashpot since the spring is not extended at this time. Evaluation of the integrals in equation (6) gives after rearrangement:

$$\frac{1}{\beta}\ln\left[\tanh\left(\frac{\beta\sigma_{ef}}{2}\right)\tanh\left(\frac{\beta\sigma_{ap}}{2}\right)\right] = -E_a K t \tag{7}$$

After rearrangement this yields the following equation:

$$\sigma_{ef} = \frac{2}{\beta}\tanh^{-1}\left[\tanh\left(\frac{\beta\sigma_{ap}}{2}\right)\exp(-E_a\beta K t)\right] \tag{8}$$

which after inserting the expression (9) for the partition of stress between the recovery and effective stress components:

$$\varepsilon_a = (\sigma_{ap} - \sigma_{ef})/E_a \tag{9}$$

yields for the anelastic strain:

$$\varepsilon_a = \frac{\sigma_{ap}}{E_a} - \left(\frac{2}{\beta E_a}\right)\tanh^{-1}\left[\tanh\left(\frac{\beta\sigma_{ap}}{2}\right)\exp(-E_a K\beta t)\right] \tag{10}$$

Equation (10) can be used to eliminate ε_a^* from equation (2) by inserting the value of ε_a when $t = t_r$, where t_r is the rupture time. Thus one obtains:

$$\left[\frac{2}{E_a}\left(R - \frac{\sigma_{ap}^2}{E_e}\right)\right]^{1/2} = \frac{\sigma_{ap}}{E_a} - \left(\frac{2}{\beta E_a}\right)\tanh^{-1}\left[\tanh\left(\frac{\beta\sigma_{ap}}{2}\right)\exp(-E_a\beta K t_r)\right] \tag{11}$$

Rearranging equation (11) gives:

$$t_r = \frac{1}{E_a K\beta}\ln\left[\tanh\left(\frac{\beta\sigma_{ap}}{2}\right)\Big/\tanh\left(\frac{\beta H}{2}\right)\right] \tag{12}$$

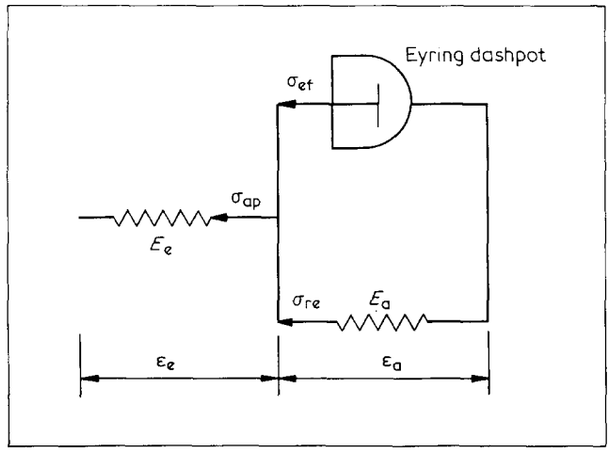

Figure 1 The three-element model used in creep rupture prediction

where $H=\sigma_{ap}-[2E_a(R-\sigma_{ap}^2/E_e)]^{1/2}$.

Equation (12) has two important consequences.

(1) It predicts the stress limit at which the specimen ruptures immediately (within 1 s) on application of load. It can be seen that t_r will be equal to zero if

$$\sigma_{ap}=(E_eR)^{1/2} \tag{13}$$

(2) It predicts the stress limit at which the specimen sustains the load indefinitely; t_r equals infinity if

$$\sigma_{ap}=\left(\frac{2R}{1/E_e+1/E_a}\right)^{1/2} \tag{14}$$

In order to investigate whether equation (12) can be used to predict the time dependence of creep rupture on the applied stress it was necessary to examine some specific data.

EXPERIMENTAL PROCEDURE

The polymer used was high-density polyethylene (HDPE) supplied by Hoechst Australia Pty Ltd and designated GA7260 ($M_w=6.1\times10^4$, $M_n=9.5\times10^3$). This was the same material as used previously[18]. It came in powder form with no additives. The HDPE was compression moulded at 180°C between induction-heated platens for 5 min after which it was either left to cool to room temperature while still in the press or water quenched to 70°C by circulating water at a rate of 2 gal/min through the heated platens. Specimens made using the first method of cooling were called air-cooled specimens. Specimens made using the second method were called water-quenched specimens. Using thin section microscopy and small-angle light scattering (SALS), the morphology of the air-cooled specimens was found to be of a degenerated spherulitic structure with an 'X' SALS pattern which was similar to that described by Maxfield and Mandelkern[19]. The water-quenched specimens were of a banded spherulitic structure with a characteristic four-leaf SALS pattern. The HDPE plaque was milled to a thickness of 2.54 ± 0.01 mm. Dumbell-shaped specimens of gauge length 20 mm and width 5 mm were made using a polymer router. The sides of the specimen were cleaned with silicon carbide polishing paper (grade 1200) to remove machinery marks and surface scratches.

Both the air-cooled and water-quenched specimens were creep rupture tested by attaching a dead weight as shown in *Figure 2* in a constant-temperature laboratory set at $25^\circ\pm1$°C. The creep rupture time was recorded automatically by placing a microswitch under the stop-rod and connecting this switch to a clock timer. The applied stress was obtained by dividing the load by the initial cross-sectional area.

RESULTS AND DISCUSSION

The air-cooled specimens all failed in a brittle mode, whereas for the water-quenched specimens ductile failures were observed at intermediate stresses, and brittle failures at high and low stresses. The dependence of failure mode on stress and morphology will be discussed in a later paper. *Figure 3* shows the plot of ln(rupture time) *versus* applied stress for the air-cooled and water-quenched samples. For the same stress level the air-cooled specimens took a longer time to rupture than the water-quenched specimens. This was attributed to the different

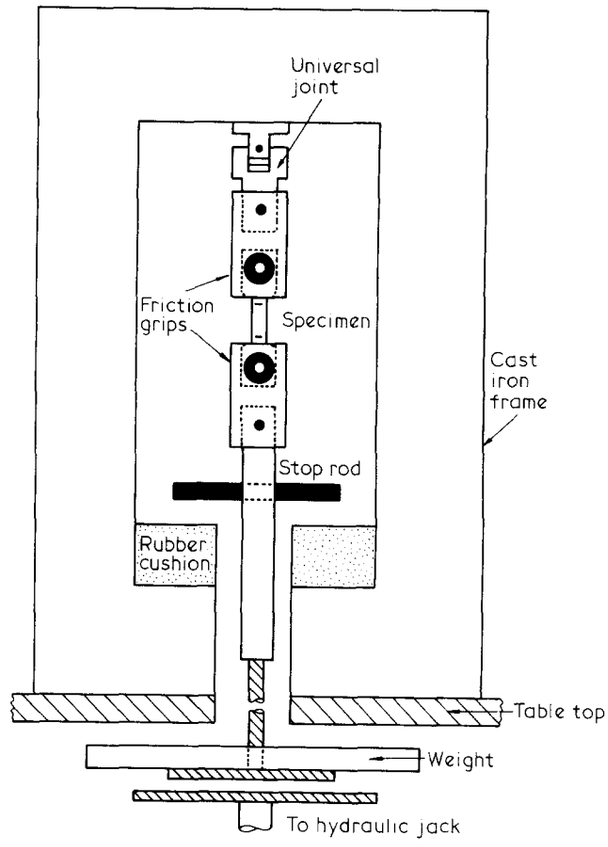

Figure 2 **Creep rupture testing equipment**

morphology of the samples, since the mean density of the (slow) air-cooled specimen was 973 kg m^{-3} and the mean density of the (fast) water-quenched specimen was 962.5 kg m^{-3}. It is interesting to note that the air-cooled and water-quenched specimens subjected to low stresses of 8 and 7.5 MPa respectively (marked by arrowheads in *Figure 3*) did not break, even after three years of creep

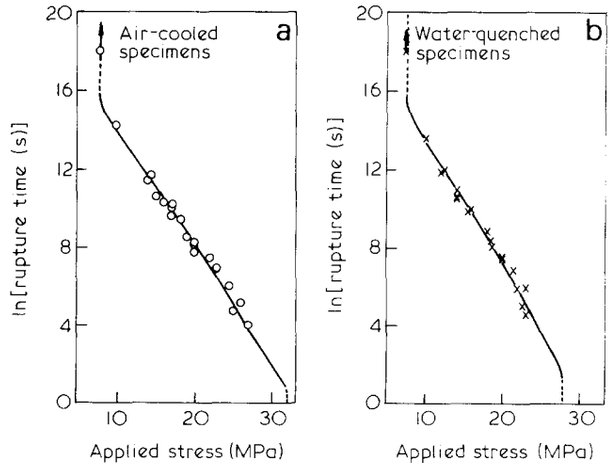

Figure 3 **Relationship between ln(rupture time) and applied stress for the (a) air-cooled and (b) water-quenched specimens (the bold lines represent the data points generated by equation (12) with the appropriate values in *Table 1*). Arrowheads indicate that the specimens are still under test**

rupture testing. These values may represent the experimental lower stress limits. The upper stress limits which were found experimentally corresponded to the stress level which gave a rupture time of about 40 s and for the air-cooled and water-quenched specimens are 27 and 23 MPa respectively. However, these values represent lower estimates of the true upper stress limits as rapid loading onto the specimen was difficult since the load was applied via a hydraulic jack. It seems likely that the true upper stress limit for rupture occurring in less than 5 s is approximately 30 and 26 MPa respectively. It should be noted that rapid straining may induce non-isothermal conditions.

The results from Crissman and Zapas[20] on creep rupture of HDPE also show upper and lower stress limits of about 32 and 7 MPa respectively. Comparing the above stress limits with Crissman and Zapas's results, it can be seen that the magnitude of the stresses are similar. Since Crissman and Zapas used HDPE of a different grade, it may be that the upper and lower stress limits of HDPE are comparatively independent of morphology. An attempt was made to fit the experimental creep rupture data of the air-cooled and water-quenched specimens using equation (12). A non-linear regression fit similar to the one used by Fotheringham and Cherry[14] was used. The estimated parameters (namely E_a, E_e, β, R and K) of equation (12) for the air-cooled and water-quenched specimens are shown in *Table 1*. The upper and lower stress limit was computed by using equations (13) and (14), respectively. The bold lines in *Figure 3* show that equation (12) describes well the relationship between ln(rupture time) and applied stress for the air-cooled and water-quenched specimens. It can be seen that the upper and lower stress limits resulting from the model corresponded well to those observed experimentally.

By comparing the cross-sectional area per chain of diamond and polyethylene, Frank[21] calculated the theoretical elastic modulus of polyethylene to be 2.8×10^5 MPa. Although the aim of this paper is not to assign the elements of the mechanical model to any particular microstructural features, it is noteworthy that the magnitude of the elastic moduli in *Table 1* approach closely the theoretical value predicted by Frank. If the elastic deformation is ascribed to the deformation of interfibrillar tie molecules, then such an identity of modulus would be expected.

Table 1 Estimated parameters of equation (12)

Parameters	Air-cooled specimens	Water-quenched specimens
E_e (MPa)	3.01×10^5	2.20×10^5
E_a (MPa)	1.00×10^4	8.00×10^3
β (MPa^{-1})	0.49	0.52
R ($MJ\,m^{-3}$)	3.19×10^{-3}	3.57×10^{-3}
K (s^{-1})	1.08×10^{-10}	1.50×10^{-10}
Upper stress limit (MPa)	31	28
Lower stress limit (MPa)	7.9	7.4

CONCLUSIONS

Using a three-element non-linear mechanical model which includes an Eyring dashpot and the Reiner–Weissenberg energy failure criterion, the creep rupture time of two types of HDPE specimens was modelled accurately. This technique also predicted the upper stress limit where the specimen sustained the load indefinitely. Both these limits were a function of the resilience, elastic and anelastic modulus of the material.

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