Time-dependent failure in poly(methyl methacrylate) and polyethylene

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Time-dependent failure of PMMA and polyethylene are characterized within the framework of a cumulative damage model for failure. It is found that the mean failure times in constant rate of stress experiments can be successfully predicted from the model using a time to fail function determined from constant stress experiments. For zero-tension sinusoidal fatigue tests, differences of up to an order of magnitude are observed between predicted and experimental failure times. PMMA and polyethylene data deviate from the predictions in different ways. In PMMA, the distribution of failure times in constant stress tests is moderately broad, as measured by the coefficient of variation, and symmetric about the mean, while in the fatigue tests the distribution is considerably broader, has a high positive skewness and shows evidence of being bimodal. For polyethylene, the distribution changes from being moderately broad and positively skewed in constant stress tests to a moderately broad, symmetric distribution in the fatigue tests. The model also predicts the total lifetime in sinusoidal fatigue tests to be independent of test frequency. Experimental results show that the lifetime of PMMA decreases with increasing frequency, although less rapidly than if the fatigue process were cycle dependent. The lifetime of polyethylene increases with increasing test frequency.

INTRODUCTION

The time-dependent failure behaviour of polymeric materials is currently a subject of great interest due to the increasing use of these materials in engineering applicatons. We have studied the failure of two polymers, poly(methyl methacrylate) (PMMA) and polyethylene, in a framework of a simple cumulative damage law which was originally proposed by Bailey[1] working with inorganic glasses. Although Bailey's s approach was purely phenomenological, special forms of this cumulative damage concept have been derived from molecular considerations by various workers including Tobolsky and Eyring[2], Coleman[3,4], and Zhurkov[5] and his coworkers[6–8].

In this work, we will examine the failure behaviour of PMMA and polyethylene experimentally using the cumulative or additive damage rule as a framework for conducting reasonable experiments; experiments which are reasonable not only in the testing of the various predictions but also in that they provide interesting information concerning failure of polymers and should serve as the beginnings of a database which would be available for the use of other workers to check and develop better models of the time-dependent failure of polymers.

FAILURE MODEL

The concept of cumulative damage is based on the assumption that material failure is the result of damage accumulation due to stress. When the damage reaches a critical value, then failure occurs. The Bailey criterion[1] and the form of the rule which we will use assumes that the rate at which damage accumulates is a function only of the current stress, σ. Then the time to fail, t_B, is related to the stress history, $\sigma(\xi)$, by the following equation:

$$\int_0^{t_B} \frac{d\xi}{\tau_B[\sigma(\xi)]} = 1 \tag{1}$$

where $\tau_B(\sigma)$ is the time to fail in constant stress experiments. Thus, each increment of time, $d\xi$, during which the load is $\sigma(\xi)$, is weighted inversely as the lifetime, $\tau_B(\sigma)$, which the specimen would have had under a constant stress, σ. In this paper we will use engineering stress throughout.

Once $\tau_B(\sigma)$ is determined from constant load (engineering stress) experiments, equation (1) can be used to predict the time to fail under any stress history. The form of $\tau_B(\sigma)$ which we found describes our data was:

$$\tau_B(\sigma) = Ae^{-B\sigma} \tag{2}$$

where A and B are constants. We note in passing that this form of $\tau_B(\sigma)$ is that which is derived from molecular considerations by Tobolsky and Eyring[2], Coleman[3,4] and Zhurkov[5]. These molecular models have been reviewed by Henderson *et al.*[9].

For the form of the time to fail function, $\tau_B(\sigma)$, given by equation (2), the time to fail, t_B, in a constant rate of stress experiment is obtained by integrating equation (1):

$$t_B = \frac{\ln(AB\dot{\sigma} + 1)}{B\dot{\sigma}} \tag{3}$$

where t_B is the time to fail and $\dot{\sigma}$ is the stress rate.

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Similarly, if a sinusoidal stress

$$\sigma(t) = p + q \sin \omega t \tag{4}$$

is applied to the sample, then the time to fail, t_B, is predicted to be:

$$t_B = \frac{\hat{t}_B(p)}{I_0(Bq)} \tag{5}$$

where $\hat{t}_B(p)$ is the time to fail at the constant stress, p; I_0 is the zero order modified Bessel function, and B is the constant from equation (2).

From an experimental point of view it is of interest to test whether or not this additivity of damage rule predicts material failure times. However, other aspects must be considered in modelling failure. In particular, there are two other points which we want to examine about the cumulative damage failure model. First, Coleman[3,4] used this form of the additivity of damage rule to describe the failure of nylon fibres. He extended the concept by using the statistics of extreme values to describe the distribution of failure times in different loading histories. His analysis predicts that the distribution of failure times is dependent on the type of loading history. Thus, the distribution of failure times in different load histories is an important parameter to examine. The other point of import is that if the rate of damage accumulation is dependent only on the stress, then the time to fail in a sinusoidal loading history is predicted to be independent of the test frequency. This is clear from equation (5).

In using the additivity of damage rule as a framework for studying failure in PMMA and polyethylene we will examine three aspects of failure.

(1) How well the average time to break predictions for specific stress histories compare with experimental results.

(2) How the statistics of failure vary with load history.

(3) How the average failure time varies with test frequency in sinusoidal load histories.

In addition, it will be of interest to compare the behaviour of the PMMA and polyethylene, not only with the model, but also with each other.

EXPERIMENTAL

Materials

A commercial grade PMMA was obtained in the form of a sheet nominally 1.5 mm thick. The sheet was cut into strips 152 mm long on a band saw. These strips were machined on a milling machine to a width of 19.0 mm and cut on a Tensil-Kut* router into dumb-bell specimens conforming to ASTM D 638 Type I in the gauge section. Specimens were conditioned in the laboratory for at least 7 days prior to testing.

The polyethylene tested was a linear high density material having a number-average molecular weight, M_n, of 15 600 and a weight-average molecular weight of 99 000[10]. The material was obtained in pellet form and moulded in a platten press at 160°C into sheets 1.0 mm thick which were then allowed to cool in the press to 90°C and subsequently removed from the mould. Dumb-bell specimens conforming to ASTM D 638 Type IV were then cut using a die and these were allowed to lie in the laboratory for a minimum of one week prior to testing.

Test conditions

The PMMA tests were conducted at 24° ± 1°C and a relative humidity of approximately 50%. The polyethylene tests were conducted at 23° ± 1°C and a relative humidity of 50 ± 5%. Each specimen's width and thickness were measured prior to testing. Mechanical testing was conducted using an Instron* servo-controlled hydraulic testing machine which was interfaced with a Hewlett–Packard* minicomputer for control and data acquisition.

The test machine is commanded by voltages generated by the computer. Constant stress experiments were conducted by applying and maintaining a single voltage for the duration of the test. Tests were conducted at a predetermined voltage so that a given engineering stress was applied throughout the test. Constant rate of stress experiments were conducted by commanding the machine from 0 to 10 volts in 0.005 volt increments. The stress at 10 volts and the time between voltage increments were chosen to give the appropriate stress rates. The sinusoidal fatigue tests were run by generating a sinewave of 200 steps per cycle for tests run at frequencies from 2×10^{-4} to 0.164 Hz. The sinewaves generated at 0.834 Hz were obtained using 40 steps per cycle. Frequencies were otherwise varied by changing the time between steps.

Time to failure was recorded for each specimen. Constant load tests on the PMMA were conducted to provide a stress range of between 55 and 75 MPa. The range of stresses for the polyethylene was 15–35 MPa. Constant rate of engineering stress tests covered a range of 0.0126 to 172.4 MPa s^{-1} for the PMMA and 10^{-3} to 10^{-1} MPa s^{-1} for the polyethylene[11]. The fatigue tests were conducted from zero to peak stresses over the same range as the dead load tests and at frequencies of from 2×10^{-3} to 0.164 Hz for the PMMA and from 2×10^{-4} to 0.834 Hz for the polyethylene. Statistical analysis was performed on the constant stress and fatigue experimental data.

RESULTS AND DISCUSSION

Additivity of damage–lifetime predictions

For constant stress tests on PMMA, the average lifetime at the stress, σ, is represented by equation (2) with $A = 4.0 \times 10^{10}$ s and $B = 0.27$ MPa^{-1}. For polyethylene, we found $A = 6.2 \times 10^7$ s and $B = 0.46$ MPa^{-1}. The results are depicted as a logarithm of the mean time to fail *versus* stress in *Figure 1*. The fit is quite good over the stress ranges covered. It is expected that at lower stresses, the form of $\tau_B(\sigma)$ would not be the same as represented by equation (2). Also, we find it interesting to note that not only does polyethylene show lower strength than PMMA, but the time to fail decreases more rapidly with increasing stress than it does for PMMA.

The first test of the applicability of the additivity of damage rule under study is from constant rate of stress experiments. Equation (3) predicts the time to fail in a constant rate of stress test. *Figure 2* shows how well the predicted values agree with those obtained experimentally for PMMA at room temperature. *Figure 3* shows results obtained by Zapas and Crissman[11] for the same polyethylene

* Certain commercial materials and equipment are identified in this paper in order to specify adequately the experimental procedure. In no case does such identification imply recommendation or endorsement by the National Bureau of Standards, nor does it imply necessarily the best available for the purpose.

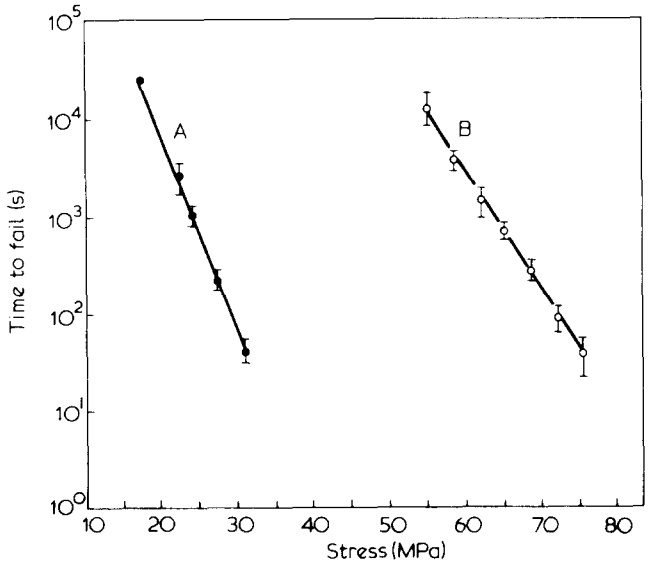

Figure 1 Time to fail vs. stress from constant stress experiments for poly(methyl methacrylate) and polyethylene. A, Polyethylene, T = 296K; B, PMMA, T = 297K

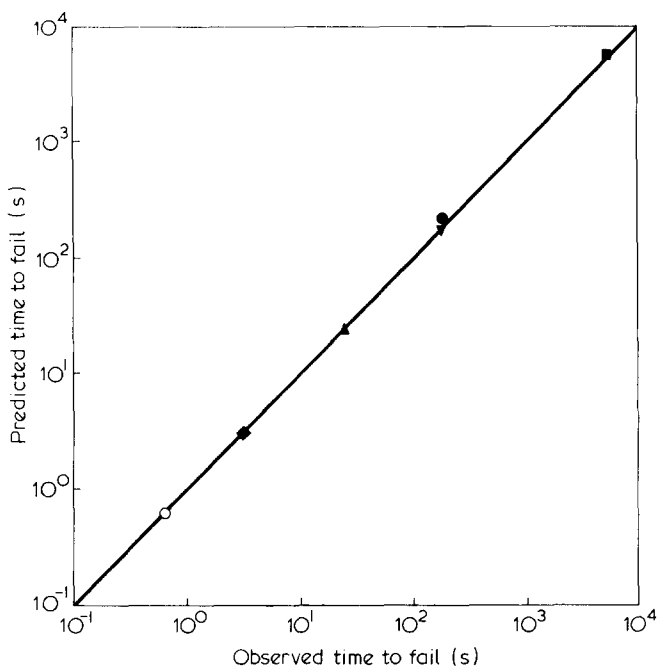

Figure 2 Time to fail predicted from additivity of damage rule vs. experimentally observed failure times in constant rate of stress experiments for poly(methyl methacrylate), T = 297K. Stress rate, $\dot{\sigma}$ (MPa s^{-1}): ○, 172.4; ◆, 32.3; ▲, 3.77; ▼, 0.471; ●, 0.404; ■, 0.0126

at two different temperatures. In both the PMMA and polyethylene, the agreement is quite good over several decades in time (stress rate). The agreement between experimental values and the theoretical values is, perhaps, not surprising. The nature of the constant rate of stress test is such that the high stresses dominate failure while the total time to reach the high stresses is what we are measuring and the bulk of the material damage occurs in a very small portion of the test time just prior to failure. Thus, this test is not a very sensitive measure of the applicability of the additivity of damage model to polymer failure.

In order to obtain more definitive information about the failure of PMMA and polyethylene, we conducted zero tension sinusoidal fatigue tests, i.e. $p = q$ in equation (4) and the peak stress is $p + q$. *Figure 4* shows the comparison of the average times to break obtained in experiments at 0.164 Hz with those predicted from equation (5) for PMMA. As can be seen, the predicted times to fail vs. peak stress form a line nearly parallel to the dead load (constant stress) data and at approximately seven-fold greater failure times. The experimental data, however, are lower than the predicted values and show a concave deviation from the predicted values. Similar, though quantitatively different results are obtained at other frequencies.

Figure 5 shows the comparison of the average times to break in experiments at 0.09 Hz with those predicted from equation (5) for polyethylene. The predicted failure times vs. peak stress form a line nearly parallel to the dead load data and at approximately five-fold longer failure times. In this case, the experimental data fall on a straight line which is higher than the predicted line and shows a greater slope. Similar, though quantitatively different results are obtained at other test frequencies.

At this point, we observe that the additivity of damage rule seems adequately to predict failure times for both PMMA and polyethylene in constant rate of stress tests. However, in zero tension sinusoidal fatigue tests, the additivity of damage rule does not successfully predict the failure times. An interesting result is that the PMMA and polyethylene fatigue lifetimes deviate from the additivity of damage predictions in greatly different ways. Deviations from predicted lifetimes are found as large as a factor of ten. While the predictions for polyethylene are conservative, those for PMMA might be dangerous if used for design purposes.

Statistics of failure

Coleman's model of failure[3,4] implies that the statistics of failure should be dependent of the nature of the applied loading history. In this section we compare the statistics of failure from constant stress tests with those for sinusoidal

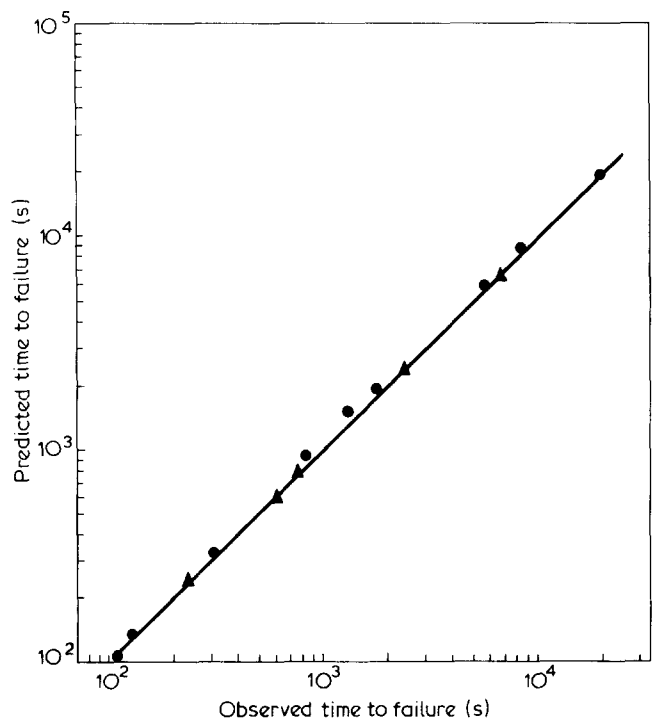

Figure 3 Time to fail predicted from additivity of damage rule vs. experimentally observed failure times in constant rate of stress experiments for polyethylene. From Crissman and Zapas[11]. Constant rate of loading: ●, 296K; ▲, 350K

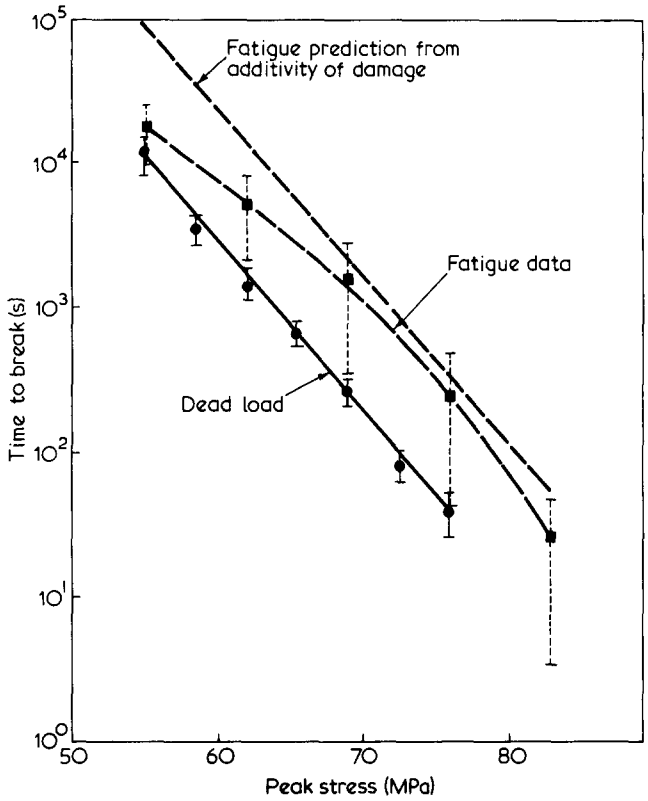

Figure 4 Comparison of failure times obtained in zero-tension sinusoidal fatigue experiments with those predicted from the additivity of damage rule for PMMA at 0.164 Hz. T = 297K

fatigue tests. Two separate measures of the distribution of failure times are used – the coefficient of variation and the moment coefficient of skewness.

The coefficient of variation is a measure of the breadth of the distribution and is defined as the standard deviation divided by the mean. The moment coefficient of skewness, γ, is a measure of the shape of the distribution, and in particular of its symmetry about the mean. It is defined as:

$$\gamma = \frac{m_3}{m_2^{3/2}} \qquad (6)$$

where the m_is are the ith moments of the distribution about the mean.

Table 1 shows the results for the constant stress tests for PMMA. *Table 2* shows the results for the sinusoidal fatigue tests on PMMA. First, we note from *Table 1* that the coefficient of variation of failure times does not seem to show any systematic variation with stress and has an approximate value of 0.30. The coefficient of skewness does not vary systematically with stress and seems to fluctuate about zero, indicating a symmetric distribution.

In the case of the fatigue data, we can see from *Table 2* that the numerical measures of the distribution of failure times are different than for the case of constant stress. Thus, the coefficient of variation appears much higher at approximately 0.60 indicating that the distribution is broader than the constant stress failure time distribution. In the case of the coefficient of skewness, there seems to be little doubt that the distribution of failure times for the fatigue tests has a positive skewness indicating that the distribution is asymmetric.

If we make the assumption that the distributions at different stresses and, for fatigue, different frequencies are the same, we can put all of the constant stress data together and all of the fatigue data together and plot histograms of frequency of failure vs. $(\tau_\beta - \bar{\tau}_\beta)/\bar{\tau}_\beta$ which normalizes each group of data to its own mean. *Figure 6* shows histograms for the constant stress data and fatigue data for PMMA. These demonstrate dramatically the change in the distribution of failure times in going from a static test to a sinusoidal fatigue test. An interesting result is also shown in the fatigue data histogram, where evidence of a bimodal distribution of failure times is apparent in addition to the skewness and breadth of the distribution.

In the case of polyethylene different results are obtained. *Table 3* tabulates the data for the constant stress tests and *Table 4* those for the fatigue tests. In examining *Table 3* we note that the coefficient of variation of failure time does not appear to vary systematically with stress level and has a value of approximately 0.25. The coefficient of skewness, on the other hand, appears to be positive, indicating a non-symmetric distribution. In the case of the sinusoidal fatigue data, we

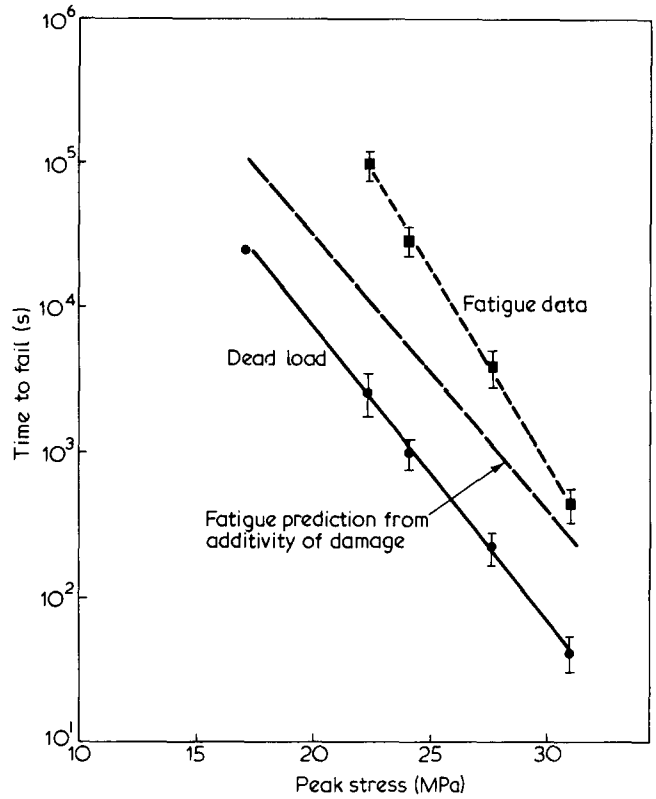

Figure 5 Comparison of failure times obtained in zero-tension sinusoidal fatigue experiments with those predicted from the additivity of damage rule for polyethylene at 0.09 Hz. T = 296K

Table 1 PMMA failure data from constant stress tests

Stress (MPa)	Number of tests	Mean failure time, (s)	Coefficient of variation of failure times	Coefficient of skewness
55.2	10	12 785	0.353	0.44
58.6	9	3551	0.233	0.14
62.1	10	1375	0.364	−0.43
65.5	10	670	0.172	−0.16
69.0	13	264	0.239	0.20
72.4	15	78.9	0.311	−0.47
75.8	30	35.5	0.456	−0.05

Table 2 PMMA failure data from zero-tension sinusoidal fatigue tests

Peak stress (MPa)	Number of tests	Mean failure time, (s)	Coefficient of variation of failure times	Coefficient of skewness
0.164 Hz				
55.2	10	18026	0.449	0.46
62.1	19	5125	0.579	0.60
69.0	17	1549	0.783	0.41
75.8	14	258	0.840	1.42
82.7	6	25.8	0.868	0.82
0.019 Hz				
55.2	10	34688	0.606	0.68
69.0	10	6386	0.509	0.02
75.8	10	630	0.721	0.58
0.002 Hz				
55.2	6	160203	0.604	0.37

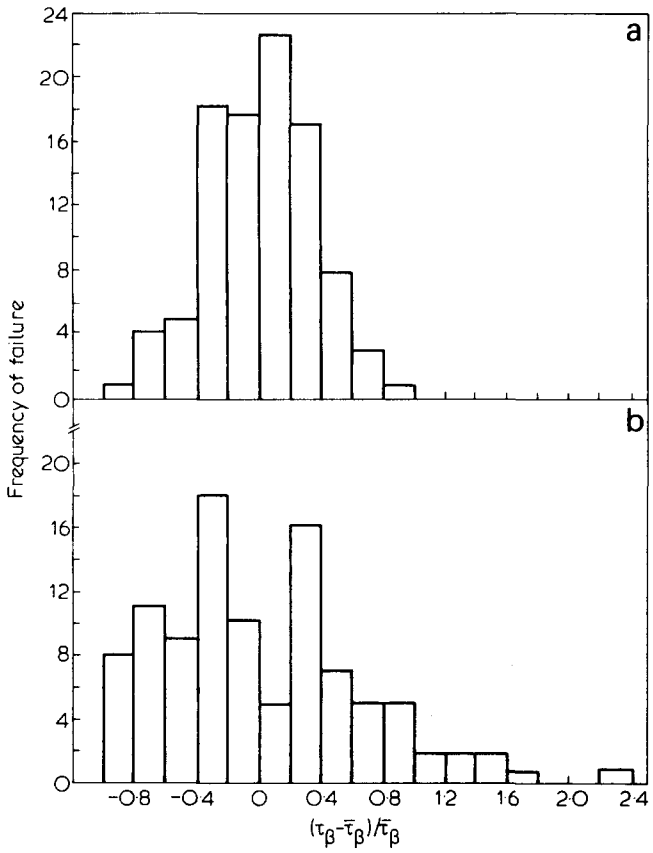

Figure 6 Frequency of failure vs. deviation from mean failure time, $(\tau_\beta - \bar{\tau}_\beta)/\bar{\tau}_\beta$ for PMMA. (a) Constant stress data, $N = 97$; (b) zero-tension sinusoidal fatigue data, $N = 102$

observe that there is no significant change with stress level in the coefficient of variation of failure times. Further, the value is approximately 0.25 as was the case for the constant stress tests. In examining the coefficient of skewness we observe, however, that it seems to be less certainly positive and that the distribution may be symmetric about the mean. Thus, there appears to be a smaller change in the failure time distributions for polyethylene in going from a static history to a dynamic load history than for PMMA.

If we again make the assumption that the distribution of failure times is independent of load or frequency, then we can again construct histograms of frequency of failure vs. $(\tau_\beta - \bar{\tau}_\beta)/\bar{\tau}_\beta$. *Figure 7* shows the comparison of the histogram in constant stress and sinusoidal fatigue testing of polyethylene. This Figure tends to indicate that the constant stress failure time distribution is somewhat positively skewed whereas the fatigue distribution of failure times appears to be more symmetric. A word of caution is needed here in noting that these data would be more conclusive if the number of tests were larger.

In summarizing this section, it appears that the distribution of failure times is dependent on the type of loading history imposed on the material. For PMMA, in going from constant stress tests to sinusoidal fatigue tests, the distribution of failure times goes from a symmetric distribution of moderate breadth to a positively-skewed, possibly bimodal distribution of considerably greater breadth. For polyethylene, the distribution seems to be somewhat positively skewed in constant stress tests and becomes more symmetric in the sinusoidal fatigue tests, while the breadth of the distribution does not change.

Effects of frequency on fatigue life

In examining how test frequency affects the lifetime of polymers in sinusoidal fatigue, the additivity of damage rule predicts that there should be no effect of frequency on life-

Table 3 Polyethylene failure data from zero-tension sinusoidal fatigue tests

Stres (MPa)	Number of tests	Mean failure time, (s)	Coefficient of variation of failure times	Coefficient of skewness
17.2	2	25193	–	–
22.4	7	2629	0.340	1.67
24.1	10	1001	0.233	1.99
27.6	10	223	0.253	0.37
31.0	10	41.7	0.285	0.34

Table 4 Polyethylene failure data from zero-tension sinusoidal fatigue tests

Peak Stress (MPa)	Number of tests	Mean failure time, (s)	Coefficient of variation of failure times	Coefficient of skewness
0.834 Hz				
22.4	5	150917	0.305	0.68
27.6	6	18241	0.251	0.70
0.09 Hz				
22.4	5	98581	0.228	0.49
24.1	8	29688	0.240	–0.42
27.6	10	3973	0.277	0.14
31.0	10	445	0.259	–0.43
34.5	6	41.1	0.349	0.21
0.01 Hz				
27.6	10	2651	0.246	0.17
0.002 Hz				
22.4	5	38177	0.199	0.24
27.6	5	1800	0.208	–0.09
2×10^{-4} Hz				
22.4	4	17838	0.233	0

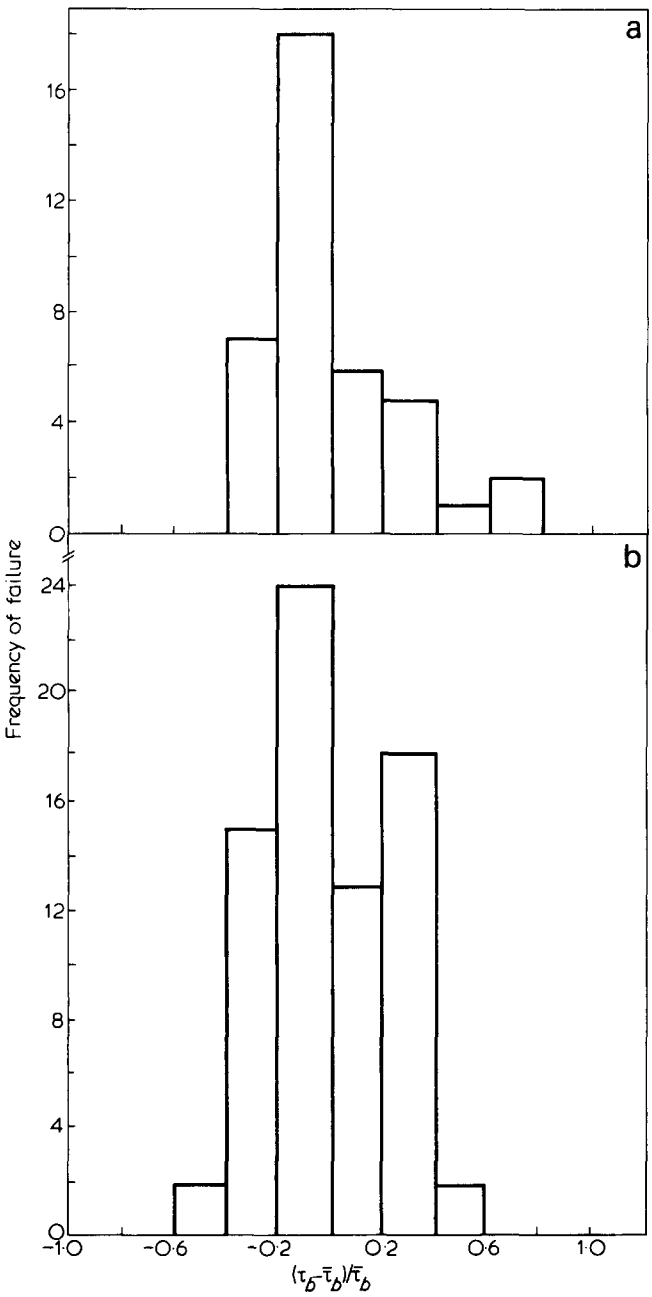

Figure 7 Frequency of failure vs. deviation from mean failure time, $(\tau_\beta - \bar{\tau}_\beta)/\bar{\tau}_\beta$ for polyethylene. (a) Constant stress data, $N = 39$; (b) zero-tension sinusoidal fatigue data. $N = 74$

time as measured by total time to failure. In contrast, many workers doing research in fatigue prefer to look at lifetime in terms of the number of cycles to failure. In this case, if failure were a cycle-dependent process then the total lifetime to failure would decrease proportionately with increasing test frequency. In presenting our fatigue data we examine time to fail relative to both additivity of damage and cycle dependent fatigue processes.

Table 2 summarized all of the fatigue test results on PMMA. Zero-tension fatigue tests were conducted at 2×10^{-3}, 0.019 and 0.164 Hz at a peak stress level of 55.2 MPa. Tests were also run at peak stresses of 69.0 and 75.8 MPa at 0.019 and 1.64 Hz. *Figure 8* shows how the fatigue lifetime changes with test frequency. The material lifetime decreases with increasing test frequency for all stress levels. Thus, the constant lifetime prediction of the additivity of damage rule is not followed by PMMA. In comparison the material behaviour is not cycle dependent but falls between the cycle-dependent fatigue and the additivity of damage predictions.

A comment is in order here concerning the possible effects of hysteretic heating on lifetime. Other researchers have observed that at relatively low frequencies and in samples with low surface to volume ratios, self-heating of the polymer can occur which results in failure by thermal processes[12–14]. However, we have conducted our tests on samples which have a high surface to volume ratio and at low frequencies. In addition, placement of a thermocouple on the surface of representative samples indicated no thermal build-up at the surface during the test. Thus, we are satisfied that we have come reasonably close to conducting an isothermal test.

In the case of polyethylene, the effects of test frequency on fatigue lifetime are markedly different from the effects observed for PMMA. We conducted tests at frequencies from 2×10^{-4} to 0.834 Hz and at stresses of 22.4 and 27.6 MPa. The results are tabulated in *Table 4*. *Figure 9* shows how the fatigue lifetime of polyethylene varies with test frequency. As can be seen, time to fail increases with increasing frequency. The way in which lifetime depends on frequency also apparently depends on the magnitude of the peak stress. It is of interest to remark that the effect of test frequency on lifetime in polyethylene not only doesn't follow the additivity of damage rule, but also runs opposite to that which would be expected from a cycle-dependent fatigue process.

In summary, the frequency dependence of fatigue processes in PMMA and polyethylene does not follow the prediction of the additivity of damage rule. Neither does a cycle-dependent fatigue process explain the effects of frequency on the lifetime of these two materials. Finally,

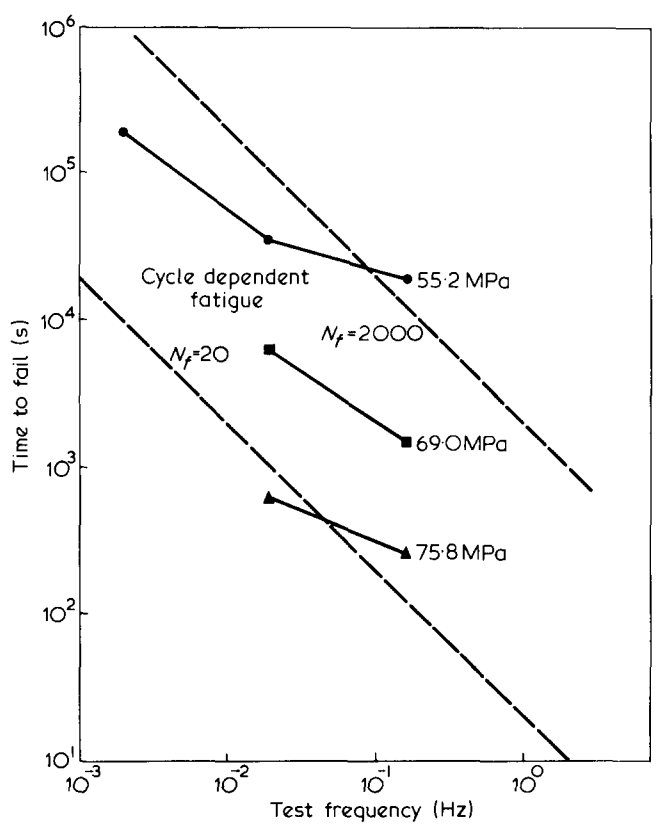

Figure 8 Time to fail vs. test frequency in zero-tension sinusoidal fatigue tests for PMMA. Broken lines represent cycle-dependent fatigue behaviour

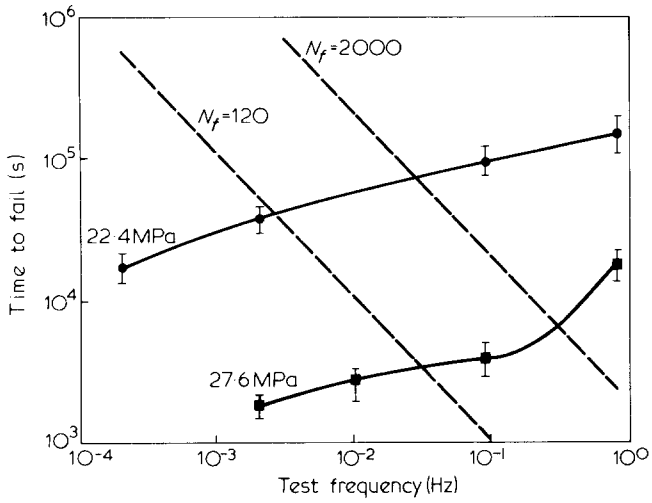

Figure 9 Time to fail vs. test frequency in zero-tension sinusoidal fatigue tests for polyethylene. Broken lines represent cycle-dependent fatigue behaviour

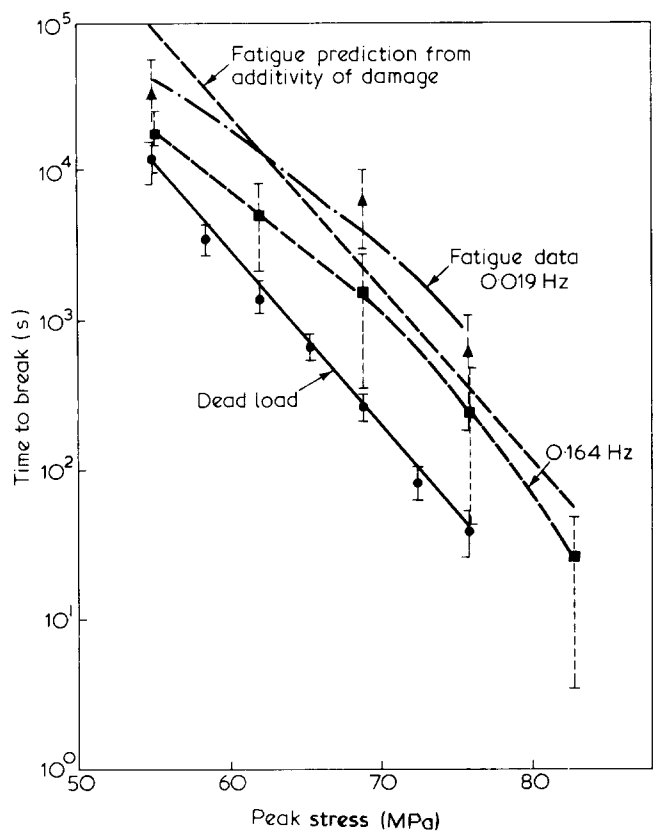

Figure 10 Summary of results of time to fail vs. peak stress for PMMA. Error bars denote one standard deviation about the mean. T = 297K

PMMA and polyethylene show opposite frequency effects on fatigue lifetimes – lifetime for PMMA decreases with increasing frequency and lifetime for polyethylene increases with increasing frequency.

CONCLUSIONS

Time to fail behaviour of poly(methyl methacrylate) and polyethylene has been characterized within the framework of an additive or cumulative damage model. The major results are summarized in *Figure 10* for PMMA and *Figure 11* for polyethylene.

In brief, the additivity of damage model allows us to predict failure times in complicated loading histories from tests to failure at constant stress. We found that the constant stress behaviour of both PMMA and polyethylene can be described by an exponential time to fail function of the form of equation (2). Times to fail in constant rate of stress tests for both PMMA and polyethylene were successfully predicted over several decades in stress rates.

In the case of zero-tension sinusoidal fatigue tests we found that the additivity of damage model was not adequate to describe the failure behaviour of either PMMA or polyethylene. In addition, the way in which the behaviour of the two materials deviated from the prediction is different. This is best seen by comparing *Figure 10* with *Figure 11*.

Two other aspects of the additivity of damage model indicated that failure models should be able to predict both the failure statistics and the effects of frequency on material lifetime. For the PMMA, the statistics of failure changed markedly from constant stress experiments to sinusoidal fatigue experiments. The error bars in *Figure 10* show the standard deviation of the measurements. The distribution of failure times in constant stress tests was symmetric as measured by the coefficient of skewness while the distribution in fatigue tests was quite positively skewed and indicated a possible bimodal nature. The distribution of failure times also broadened considerably as measured by an approximate doubling in the coefficient of variation in going from the constant stress to sinusoidal fatigue tests in PMMA.

The statistics of failure for the polyethylene did not change as dramatically as they did for PMMA. The breadth of the distribution as measured by the coefficient of varia-

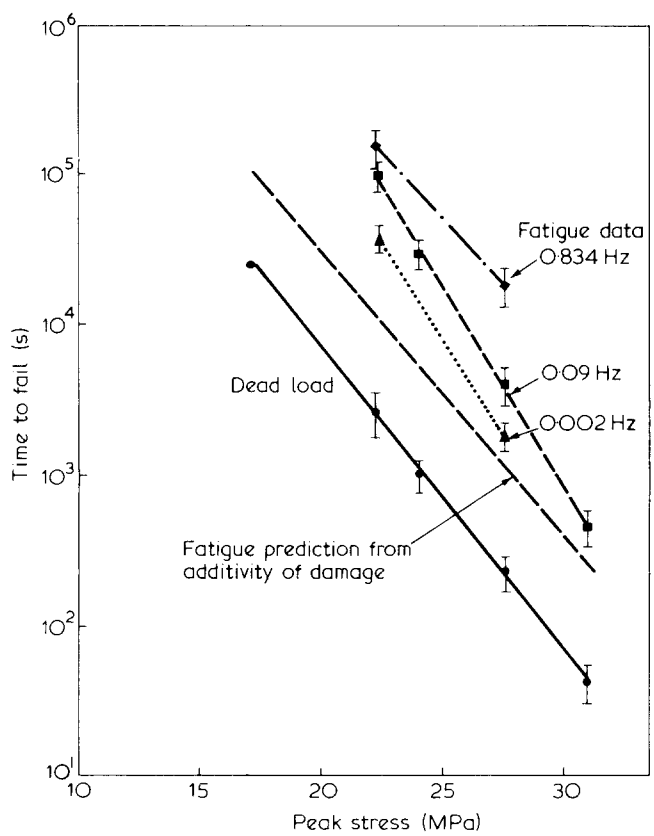

Figure 11 Summary of results of time to fail vs. peak stress for polyethylene. Error bars denote one standard deviation about the mean. T = 296K

tion did not change significantly. However, there was a change from a somewhat positively-skewed distribution of failure times in the constant stress results to a symmetric distribution in the fatigue experiments.

In fatigue experiments, the additivity of damage model predicts that the total failure time is independent of the test frequency. In the case of PMMA we found that the material lifetime decreased with increasing frequency. It did not decrease in a fashion which would be consistent with a cycle-dependent fatigue model. Also the frequency sensitivity is not consistent with results reported in fatigue crack propagation tests[15,16]. The fatigue lifetime of polyethylene was found to increase with increasing test frequency.

The type of experimental data which we have presented here should serve to help extend or modify existing theories such as the additivity of damage model. In addition, it should prove useful to other workers to check other models which have been developed for describing time-dependent failure of materials.

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