A Model for Service Life of Polyethylene Pipe Exhibiting Ductile–Brittle Transition in Failure Mode*

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Synopsis

This article proposes a model for predicting failure time of stressed polyethylene pipe materials that exhibit a failure mode transition from brittle to ductile as stress is increased. The model is based on data obtained using the constant tensile load (CTL) test and takes into account stress-versus-failure-time behavior in both brittle and ductile regimes, as well as in the transition regime. The model permits quantification of the ductile–brittle transition behavior not only from the standpoint of the location of the transition but also its breadth. It is illustrated that knowledge of these two separate parameters opens new avenues for understanding the molecular basis of the transition process. This research was conducted under the sponsorship of the Gas Research Institute.

INTRODUCTION

The use of the ductile–brittle (D–B) transition as an indicator of failure in stressed polymeric materials is well known. This phenomenon for polyethylene can be illustrated by Figure 1, which shows time to failure versus stress. In the low-stress regime where failure times are long, failure is governed by crack growth and the failures obtained are referred to as brittle failures because of the appearance of the fracture surface. In the high-stress/short-failure time regime, failure results from creep rupture due to the viscoelastic nature of the material. As has been pointed out by Hoffman,[1] "a fundamental understanding of both of these modes of failure is necessary to better estimate long-term behavior of polymers in service." It is also important, however, to understand the factors that govern the transition region, namely its sharpness, as well as its location in stress-time coordinates. Furthermore, to gain such understanding, it is useful to be able to mathematically model the relationship between failure time and stress in the transition regime.

In this report, stress-versus-failure-time data for polyethylene in air and in a surfactant solution[†] are addressed. The test data consist of failure times for polyethylene pipe specimens which were subjected to the constant tensile load (CTL) test until failure occurred. Environments were air or a surfactant solution, and temperatures were 23 and 35°C. Both the surfactant solution and the increased temperature served to accelerate the failure processes.

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[†] 1% Nonylphenoxypoly(ethyleneoxy)ethanol (Igepal CO-630).

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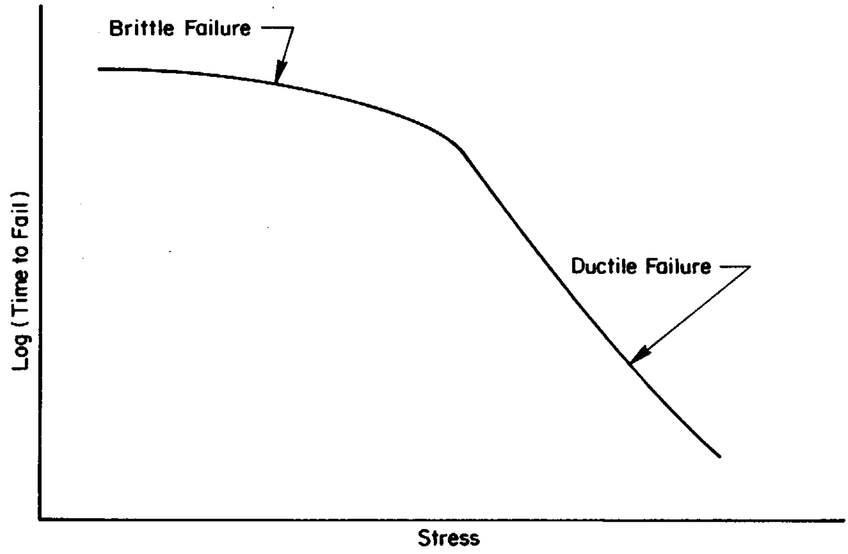

Fig. 1. Typical stress-versus-failure-time curve.

Typically, such stress-versus-failure-time data are treated by modeling the two regimes separately. The location of the ductile–brittle transition is then taken as the point of intersection of the two separate equations. However, this approach is problematical for two reasons. First, it sometimes forces one to make arbitrary decisions as to whether a datum is in the ductile or brittle regime. When this choice is difficult, the datum in question is often omitted—thereby reducing the information content of the research program and possibly biasing the fitted equations. Second, this approach does not account for the broadness of the ductile–brittle transition region. In order to understand the factors that govern this transition, it is useful to be able to quantify not only the location of the transition regime but also its breadth. This paper proposes a single model for fitting data in both low-stress (brittle) and high-stress (ductile) regimes, as well as in the transition regime. This is achieved by joining the two separate models by a stochastic element that addresses the probability of one or another of the failure modes being operative. The parameters of the model permit improved characterization of the transition regime.

STOCHASTICALLY SEGMENTED REGRESSION MODEL

Let the brittle and the ductile stress-versus-failure-time segments be linearly (in log time) related as follows:

$$\log(t_b) = B_o - B_1 S \tag{1}$$

and

$$\log(t_d) = D_o - D_1 S \tag{2}$$

where B_1 and D_1 are the slopes of the stress-versus-failure-time data in the brittle and ductile stress regions, respectively, B_o and D_o are the corresponding intercepts, S represents stress, and t represents failure time. For the special case of isothermal testing, these equations are consistent with the relationship proposed independently by Zhurkov[2] and by Kelen.[3]

Finally, let P represent the probability of failure by the high-stress (ductile) failure mode. For very low stresses, P would be nearly zero and for very high stresses P would be nearly 1.0. At a stress for which the probability of failure by either failure mode is equal, it is reasonable for P to be near 0.5—depending on the distribution selected for P.

Equations (1) and (2) can then be combined as follows:

$$\log(t) = (1 - P)[B_o - B_1 S] + P[D_o - D_1 S] \tag{3}$$

This equation states that, for any stress, the failure time is a weighted average of that predicted for Eqs. (1) and (2) with the weighting factors being the respective probabilities of the two failure modes. At stress levels far from the transition region, Eq. (3) reverts essentially to Eq. (1) or Eq. (2), since P will be nearly 1.0 or nearly 0.

This concept of joining two deterministic line segments by a stochastic element was first described by Quandt,[4] who applied the concept to econometric data.

SELECTION OF AN APPROPRIATE PROBABILITY DENSITY FUNCTION (PDF)

Ideally, the choice of a PDF will be based upon a mechanistic understanding of the underlying phenomena. Unfortunately, our present understanding of the ductile–brittle transition process is insufficient to make such judgments. Therefore, we must resort to statistical devices and use a PDF that provides the "best" fit to the data. However, for such diagnosis to be made, it is necessary to have a considerable amount of data in the transition region. Lacking this, it has been found that several choices appear consistent with the data. For example, Derringer[5] has employed both Weibull and normal distributions for polyethylene pipe stress rupture data with equal success. Therefore, it is reasonable to base the choice on computational ease—so long as the fit to the data is adequate and the PDF selected does not conflict with theoretical considerations.

For the stress-versus-failure-time data considered in this study, Weibull, normal, and (smallest) extreme value (Type 1) distributions were all considered. Each appeared adequate to describe the data, but the latter resulted in faster convergence of the nonlinear regression algorithm being used. The Type 1 extreme value distribution function over stress is represented as follows:

$$P = 1 - \exp\{-\exp[(S - \mu)/\sigma]\} \tag{4}$$

Here S represents stress, μ is the location parameter, and σ is the scale parameter. μ is related to where the transition occurs, whereas σ is a measure of the breadth of the ductile–brittle transition region.

APPLICATION OF MODEL

Figures 2, 3, and 4 show fits of this model to stress-versus-failure-time data for three different types of polyethylene pipe and/or testing environments. In all cases, the fits were accomplished using nonlinear regression analysis which employed a pattern search algorithm for minimizing residual sums of squares about the fitted equation. Figure 2 is for CTL data from 2-inch polyethylene pipe tested in a surfactant solution at 35°C. Figure 3 is for CTL data from a 2-inch polyethylene pipe tested in air at 23°C, and Figure 4 is for CTL data from a 4-inch polyethylene pipe also tested in air at 23°C. None of the three materials were the same, although they were all medium-density polyethylenes. The corresponding data are presented in Tables I, II, and III, respectively. Finally, the parameters for fits of Eq. (3) are presented in Table IV along with their respective standard errors of regression.

The μ and σ parameters from Eq. (3) can be used to define a ductile–brittle transition point as well as a D–B transition region. A logical choice for the D–B transition point is that ordered pair (S', t') such that at a stress, S' there is an equal probability of obtaining a ductile or a brittle failure. S' is the median of the extreme value distribution and is calculated as follows:

$$S' = \mu - 0.3665(\sigma) \tag{5}$$

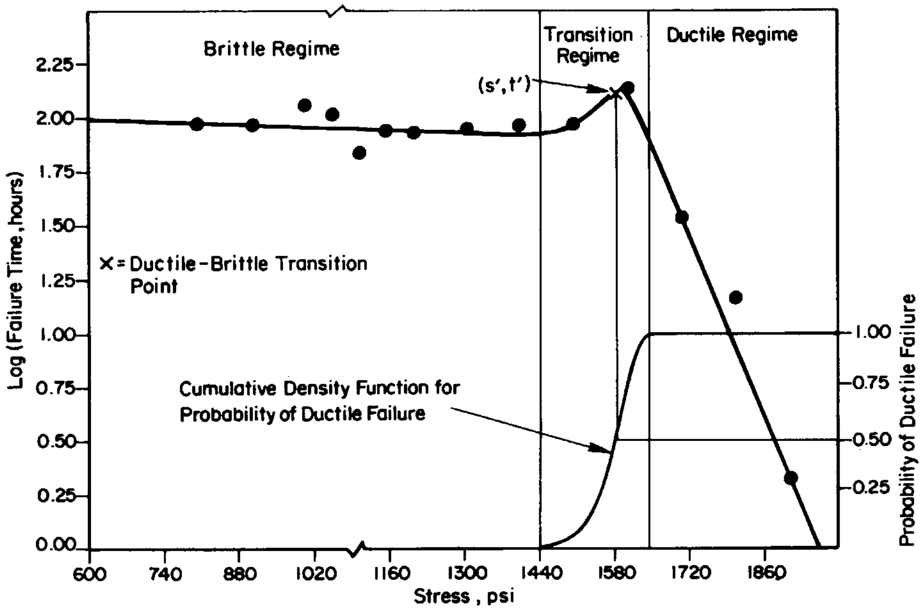

Fig. 2. Equation (3) fitted to CTL data for 2-in. polyethylene pipe specimens tested in surfactant solution at 35°C.

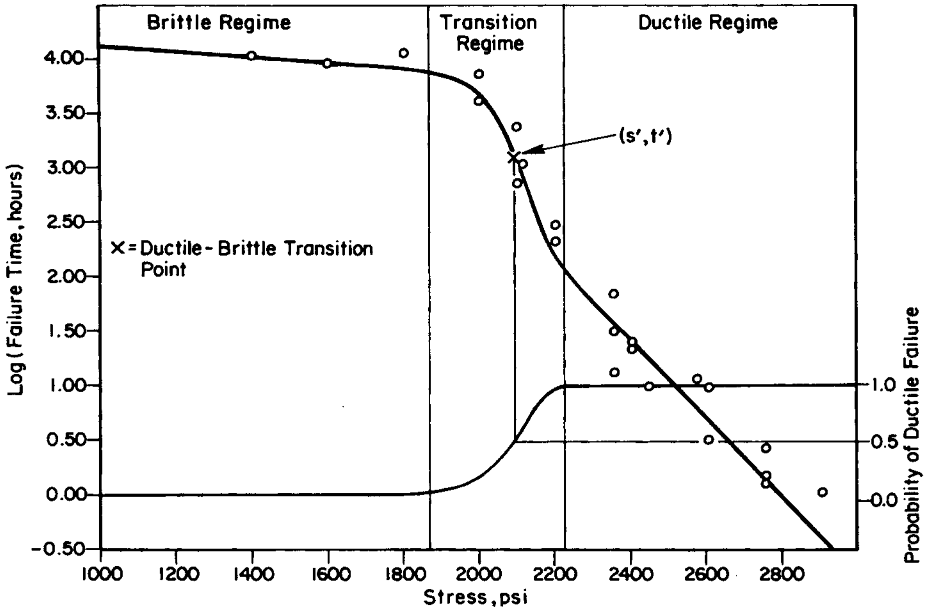

Fig. 3. Equation (3) fitted to CTL data for 2-in. polyethylene pipe specimens tested in air at 23°C.

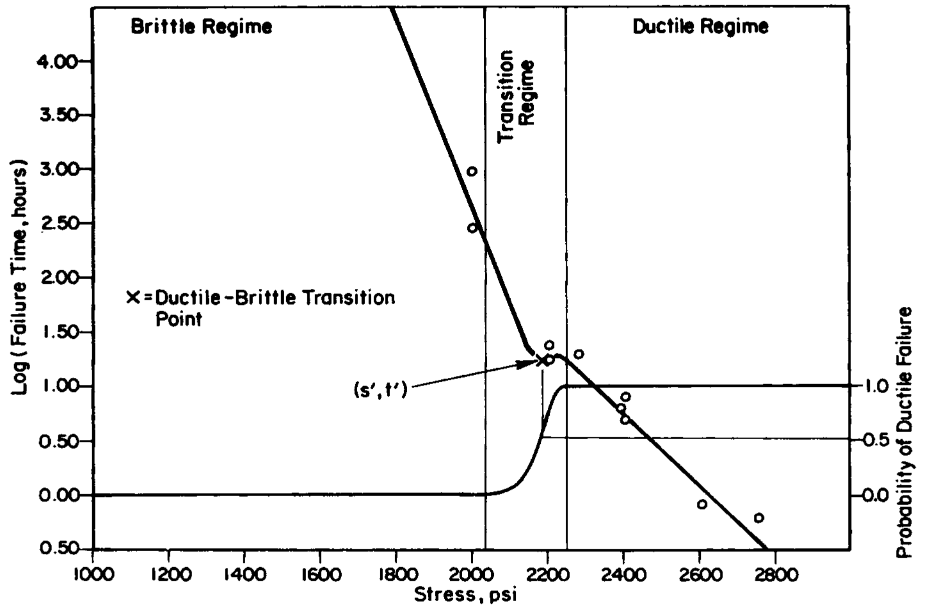

Fig. 4. Equation (3) fitted to CTL data for 4-in. polyethylene pipe specimens tested in air at 23°C.

TABLE I

CTL Test Data for 2-in. Polyethylene Pipe Specimens Tested in Surfactant Solution at 35°C (Fig. 2)

Stress (psi)	Failure time (h)
800	91.59
900	89.57
1000	112.43
1050	101.04
1100	67.34
1150	84.85
1200	82.91
1300	85.56
1400	90.06
1500	89.41
1600	130.2
1700	33.36
1800	14.17
1900	2.04

TABLE II

CTL Test Data for 2-in. Polyethylene Pipe Specimens Tested in Air at 23°C[a]

Stress (psi)	Failure time (h)
2900	1.0
2750	1.2
2750	1.4
2600	9.0
2598	3.0
2570	10.7
2442	9.2
2400	19.7
2400	22.8
2353	12.2
2353	29.3
2353	65.1
2200	195.8
2200	275.5
2113	1024.0
2100	680.1
2100	2227.2
2000	6755.1
2000	3867.1
1800	10696.4
1600	8660.9
1400	1006.7

[a] Courtesy of Argonne National Laboratory.

TABLE III
CTL Test Data for 4-in. Polyethylene Pipe Specimens Tested in Air at 23°C[a]

Stress (psi)	Failure time (h)
2750	0.58
2601	0.78
2400	4.60
2400	7.60
2389	5.88
2277	18.60
2200	16.58
2200	16.80
2200	22.38
2000	271.50
2000	894.49

[a] Courtesy of Argonne National Laboratory.

TABLE IV
Fitted Parameters for Fits of Eq. (3) to Three CTL Data Sets

Material	Parameters						Standard deviation
	μ	σ	B_o	$B_1 \times 10^{-4}$	D_o	$D_1 \times 10^{-3}$	
2-in pipe, surfactant solution 35°C (Fig. 2)	1595	32.3	2.048	0.920	11.926	6.073	0.094
2-in pipe, air, 23°C (Fig. 3)	2152	114.7	4.017	0.00	8.169	2.864	0.213
4-in pipe, air, 23°C (Fig. 4)	2194	32.28	20.89	91.00	8.733	3.318	0.222

The corresponding value of t' can be calculated by substitution of S' into Eq. (3). Calculated values for the three materials under discussion are presented in Table V.

It is also useful to demarcate a D–B transition regime. Such a regime will be defined as all stresses for which probability of either brittle or ductile failure is greater than 1%. These transition regimes are illustrated in Figures 2–4, along with the brittle and ductile regimes. In these figures the D–B transition points are designated by (S', t').

TABLE V
CTL Ductile–Brittle Transition Points for All Three Materials

Material	Ductile–Brittle Transition Stress and Time	
	Stress, S' (psi)	Time, t' (h)
2-in, Igepal, 35°C	1583	127.8
2-in, air, 23°C	2110	1180.0
4-in, air, 23°C	2182	18.3

RELATION OF S' AND D–B TRANSITION REGIME TO OTHER POLYMER PROPERTIES

It is reasonable to assume that the breadth of the D–B transition is as important a parameter as the location of the D–B transition point itself. Having a tool for objectively quantifying these separate phenomena, it is useful to explore their relationships to fundamental polymer properties. This was done for a series of different medium-density polyethylene pipe samples. In this series, a number of polymer characterization tests were run, in addition to the fits of failure time data to Eq. (3). All of the test specimens were

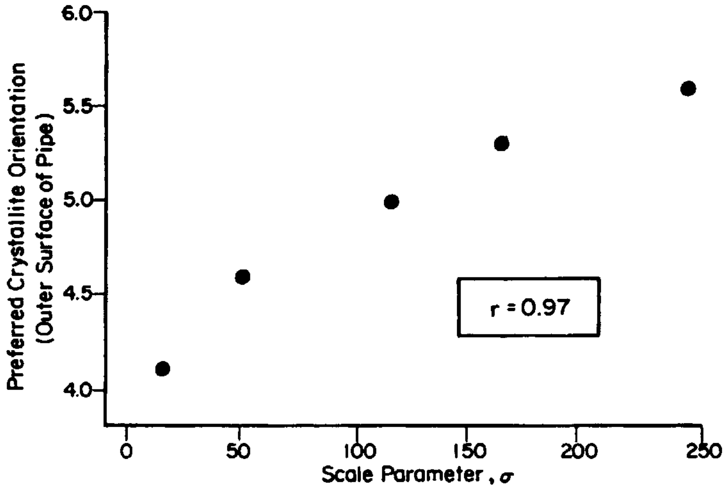

Fig. 5. Extreme value scale parameter σ [from Eq. (3)] plotted versus preferred crystallite orientation. $r = 0.97$.

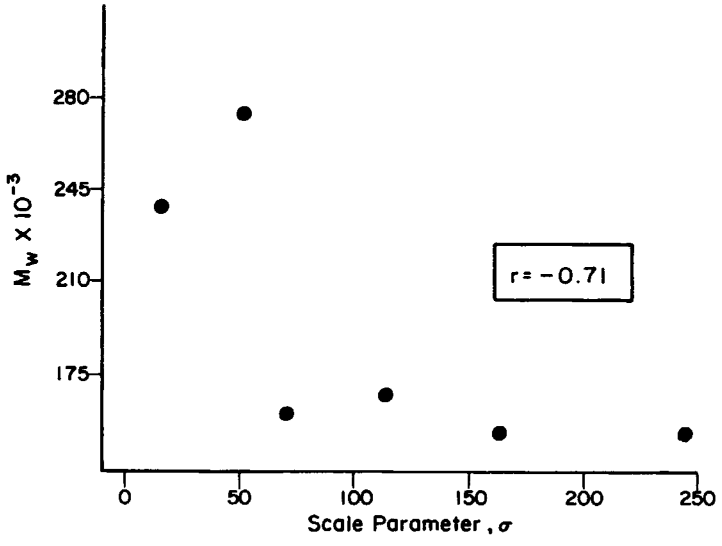

Fig. 6. Extreme value scale parameter σ [from Eq. (3)] plotted versus weight-average molecular weight. $r = 0.71$.

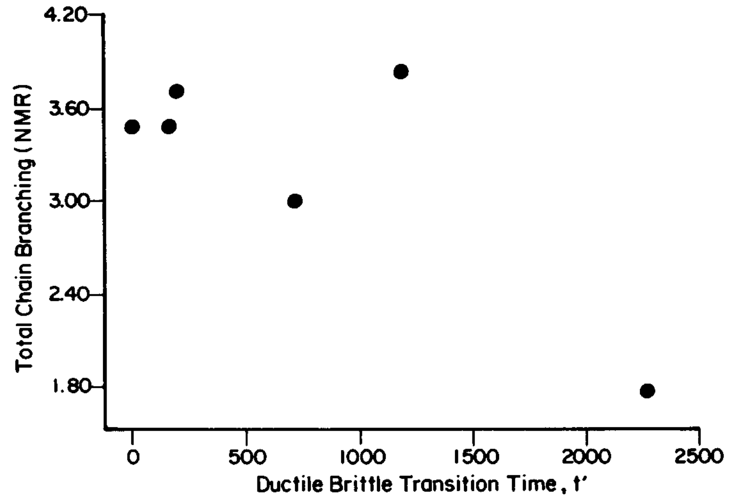

Fig. 7. Ductile-brittle transition time, t' plotted versus total (NMR) chain branching. $r = 0.79$.

randomly selected so as not to introduce bias. Some of the more interesting relationships are shown in Figures 5–7. All of these relationships are statistically significant at the 5% level of significance, or lower. Figure 5 shows the scale parameter plotted versus preferred crystallite orientation as measured by x-ray diffraction. Increased values were associated with increased levels of orientation. The results fall on a smooth curve and exhibit a linear correlation coefficient of 0.97. Figure 6 shows σ plotted versus weight-average molecular weight. It appears that increasingly broad D–B regimes are associated with lower molecular weights. The correlation coefficient in this case was -0.71. In Figure 7, the D–B transition time, t', is plotted versus total chain branching as measured by nuclear magnetic resonance. Here, increased transition times are associated with lower levels of chain branching. The correlation coefficient is -0.79. Causation, of course, cannot be attributed to any of the above associations; they are simply suggestive of areas which may prove fruitful for future investigations.

SAMPLE SIZE

Sample size requirements for fitting the model proposed in this paper for various levels of precision is easily the subject for a separate report. For example, some of the factors that dictate sample size are the scatter in the data, the sharpness of the ductile–brittle transition, the slopes of the brittle and ductile failure lines and the hypothesis being tested. However, it is possible to make some rough estimates of sample size by treating the three regimes of the curve separately. First, since each of the three segments of the curve is estimated by two parameters it would make sense to have equal numbers of specimens in each of the three stress regions. However, since the ductile–brittle transition can occur over a small stress interval that is not known in advance, it would be sensible to allocate more specimens to intermediate stresses than to the high and low stress regions. This is especially the case if the major focus of the study is to estimate the ductile–brittle transition behavior. A rough minimum sample size might be 15 specimens run at 12 different stress levels with 3 each at the low and high stress conditions and 6 spread over the intermediate stress regions. In addition, duplicate determinations for estimating model inadequacy are recommended for at least one stress level in each stress regime, bringing the total number to 15 specimens. This will give six degrees of freedom for fitting the model, six for lack of fit, and three for pure error estimation. This degree of freedom allocation is sufficient to permit a reasonable test for model adequacy. It will be noted that the data for Figure 4 do not meet these guidelines. These data, however, were all that were available and were included in order to illustrate the unusual case where the brittle regime exhibited a steeper slope than that for the ductile regime. It is certainly desirable, however, to generate additional data to confirm such unusual behavior.

CONCLUSIONS

A model has been proposed for polymer stress-versus-failure-time data exhibiting two distinct failure modes. The model accounts for behavior in three regimes: brittle failure, ductile failure, and transition. A particularly useful aspect of the model is that the ductile–brittle transition is characterized by both a location and a scale parameter. The former dictates where the transition occurs, whereas the latter dictates how rapidly (over a range of stress) it takes place. These two parameters can be related separately to fundamental polymer physical properties.

Prediction of the transition location has been done heretofore in a crude manner by finding the intersection of the two straight line segments. Estimation of the breadth of the transition, however, has not been done, but is made possible by the fit of the proposed semimechanistic model. This model, therefore, permits more information to be extracted from the data than has heretofore been possible.

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