The Time Dependence of Surface Energy in Cohesive Fracture

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Synopsis

The time-temperature dependence of the cohesive fracture energy is deduced from experiments on a centrally cracked sheet of butadiene-acrylonitrile-acrylic acid viscoelastic terpolymer crosslinked with an epoxy curing agent. Analytic results based upon a cylindrical flaw model of the crack permit the segregation of the fracture energy time dependence from that of the relaxation modulus.

INTRODUCTION

The exact analysis of cohesive fracture in a material whose properties are time and rate dependent is, as a practical matter, almost impossible, especially for an arbitrary geometry with a general loading. On the other hand, there are important design considerations which require the calculation of time and temperature-dependent fracture. Because the analytical determination of the fracture threshold is prohibitive for the general case, it was considered desirable to explore the possibility of modeling some representative flaw for which the essential features of the time dependency would be preserved, thus leading at least to a phenomenologically correct expression of the fracture. To this end, Williams[1] proposed a spherical, or cylindrical, flaw geometry in an incompressible, linear viscoelastic material subjected to uniform tension far from the flaw. By an application of the thermodynamic power balance equation, which is essentially the time-dependent form of the classic Griffith formulation, it was possible to deduce analytical results which appeared reasonable and, as might be expected, dependent upon the loading history. Subsequent applications of this idea for isothermal fatigue loading[2] also led to results which were qualitatively in agreement with experimental results and even correlated well with microscopic failure measurements in terms of free radical production in nylon using electron paramagnetic resonance.[3]

In principle then it was demonstrated that the Griffith energy balance approach could be extended to at least one realistic case of time-dependent fracture, specifically in an incompressible linearly viscoelastic material. For practical purposes, even as a temporary expedient, it was proposed that the time dependency factor deduced in this special problem could be used to

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multiply the elastic criteria obtained for sharp-edged cracks and for any general loading.

To illustrate this point, the well-known Griffith result for through crack length $2c$ in an elastic sheet of infinite extent subjected to equal biaxial loading gives the critical applied stress at fracture, σ_{cr}, as

$$\sigma_{cr} = \sqrt{\frac{2}{\pi} \frac{E\gamma_c}{c}} = \sqrt{\frac{2}{\pi}} \sqrt{\frac{\gamma_c/c}{D}} \tag{1}$$

where γ_c is the energy required to create new cohesive fracture surface and D is the linear elastic tensile compliance (inverse modulus, E). By way of comparison, it was found that the critical stress for a through cylindrical flaw of diameter $2a$ in the same infinite sheet subjected to step-applied equal biaxial tension in a linear viscoelastic but incompressible material was

$$\sigma_{cr} = \sqrt{\frac{2}{4}} \sqrt{\frac{\gamma_c/a}{2D_{crp}(t) - D_g}} \tag{2}$$

in which $D_{crp}(t)$ is the tensile creep compliance having the limit (glassy) value D_g at vanishingly small times, $D_{crp}(0) \equiv D_g$. Comparison of eqs. (1) and (2) readily shows the numerical similarity of the results in the glassy elastic region where they should be similar, i.e., in the ratio of $\sqrt{2/\pi}$: $\sqrt{2}/4 = 1.14$, and that the time dependency for this particular loading is reflected by replacing the elastic compliance D by $2D_{cr}(t) - D_g$.

Thus, in the absence of a better theory, one speculates that the time-dependent fracture criterion for a *crack* in a linear viscoelastic material might be closely represented, for the step loading, by

$$\sigma_{cr} \simeq \sqrt{\frac{2}{\pi}} \sqrt{\frac{\gamma_c/c}{2D_{crp}(t) - D_g}} . \tag{3}$$

While this type of correlation is encouraging, it is important to recognize and emphasize certain shortcomings as described in the original paper.[1] First of all, there is the obvious caution related to the incompressibility assumed in the model material for analytic simplicity, which was invoked in the Griffith case. Second, the time dependency factor was deduced for a uniform loading condition only, again for analytic simplicity, and some changes might be expected for a general triaxial loading, especially if imposed in conjunction with a compressible material. Third, the flaw growth in the model was assumed to be circularly or spherically symmetric, which is frequently questionable, particularly for the flaw size history after initiation.[4] One should recall, however, that both Griffith[5] and Sneddon[6] also assumed symmetric and simultaneous crack extension in their work on two-dimensional and penny-shaped cracks, respectively.

The fourth assumption which was incorporated in the original work is that to which the main point of this paper is directed. The derivations of all the previous results were based upon a time-independent value of the cohesive

fracture energy γ_c. As pointed out at the time, this assumption was imposed primarily as a matter of analytic convenience, plus an apparent uncertainty or disagreement among physical chemists as to the basic microscopic behavior being represented. Without delving at this time into the rationale emerging over the past few years, it may be remarked that a rather extensive series of fracture experiments have now been completed. They show that the value of γ_c is not only time dependent, but also time-temperature dependent in the WLF-shift factor sense.[7]

The appropriate extension of the basic theory is presented along with an analysis of the experimental data.

THEORETICAL ASPECTS

For the reasonably general case of fracture, the thermodynamic power equation can be written as

$$\dot{I} = \dot{F} + 2D + \dot{K} + \dot{S} \qquad (4)$$

which states that the rate of work input $\dot{I}$ to the system must equal the rate of storing free energy $\dot{F}$ and dissipation $2D$ plus the rates of converting kinetic energy $\dot{K}$ and surface energy $\dot{S}$. Specifically, using the notation of Sokolnikoff,[8] one has

$$\dot{I} = \int_A \overset{v}{T_i}\, \dot{u}_i\, dA \qquad (5)$$

$$\dot{F} + 2D = \frac{d}{dt} \int_V \int_0^t \sigma_i\, \dot{\epsilon}_i\, dt\, dV \qquad (6)$$

$$\dot{K} = \frac{d}{dt} \int_V \int_0^t \rho \dot{u}_i \ddot{u}_i\, dt\, dV \qquad (7)$$

$$\dot{S} = \frac{d}{dt} \int_A \gamma_c(t) dA \qquad (8)$$

where $A(t)$ and $V(t)$ indicate the surface area and volume, respectively, both of which are time dependent during crack growth after fracture initiation.

In the previous analysis, as indicated earlier, it was convenient to assume that γ_c was not a time-dependent function. It turns out, however, that if it is, the time rate of change of the energy to create new cohesive fracture surface is, from eq. (8),

$$\dot{S} = \gamma(t) dA/dt. \qquad (9)$$

The incorporation of this flexibility into the analysis for either a cylindrical or spherical flaw in a linearly elastic, incompressible medium subjected to uniform tension at infinity leads to a very simple modification of the earlier results. Specifically, one merely replaces in the former deductions the constant value of γ_c by its time-dependent function $\gamma_c(t)$. (In the former paper[1] the subscript c was omitted. Since then similar results have been

developed dealing with adhesive fracture.[9]   It has become necessary therefore to distinguish between the energies to create new surfaces in these phenomenologically different situations represented by γ_c cohesive, and γ_a, adhesive, fracture.)

For example, if a prescribed displacement, $u(b,t) \equiv u_0 g(t)$ is applied at a large distance from the flaw ($a \ll b \to \infty$), the thermodynamic criticality condition, in terms of the relaxation modulus $E_{rel}(t)$, becomes

$$\dot{a} \left\{ 2a\gamma_c(t) - \frac{4b^6}{a^4} \left(\frac{u_0}{b}\right)^2 \int_0^t \frac{\partial g}{\partial \xi} \right.$$
$$\times \left[E_g g\ (\xi) + \int_0^\xi \frac{\partial E_{rel}(\xi-\tau)}{\partial(\xi-\tau)}\ g(\tau)d\tau \right] d\xi - \frac{\rho}{2}\ u_0\ \frac{b^4}{a^2}\ \dot{g}^2\ (t) \right\} = 0 \quad (10)$$

where the solution $\dot{a}(t) = 0$ corresponds to the initiation (zero growth) condition before fracture, and the vanishing of the bracketed term is the desired time-dependent fracture condition for determining $a(t)$.

Constant Strain Rate Loading.   For the special case in which $g(t) = c't$, i.e., $u(b,t) = u_0 c't$, the fracture strain at the flaw, neglecting the kinetic energy,

$$\epsilon_\theta^2(a,t) = \left[\left(\frac{b}{a}\right)^3 \epsilon_\theta(b,t) \right]^2 = \frac{\gamma_c/a}{2t^{-2}E_{rel}^{(2)}(t)} \quad (11)$$

in which $E_{rel}^{(2)}(t)$ is the second integral of the relaxation modulus up to the time t.   In particular, at fracture $t = t_0$, $a(t_0) \equiv a_0$, there is the explicit result

$$\epsilon_\theta(a_0,t_0) = \sqrt{\frac{\gamma_c/a_0}{2t^{-2}E_{rel}^{(2)}(t_0)}} \quad (12)$$

with

$$E_{rel}^{(2)}(t_0) = \int_0^{t_0} \int_0^\xi E_{rel}(\xi)d\xi d\tau. \quad (13)$$

Suppose it were possible to conduct an actual experiment upon this special spherical flaw configuration having a constant initial flaw radius a_0. One would then predict for the previous case, in which a constant γ_c was assumed, that if the experimentally measured failure strains $\epsilon_\theta(a_0,t_0)$ were multiplied by the square root of the weighted, $2t_0^{-2}$, second integral of the relaxation modulus, a constant value $(\gamma_c/a_0)^{1/2}$ would result independent of the time of failure t_0.   On the other hand, if $(\gamma_c/a_0)^{1/2}$ did not experimentally result in a constant, one would infer that γ_c was (failure) time dependent. In principle, this experiment was performed.   Because it was impossible to duplicate the spherical flaw experimentally, however, it was assumed that the viscoelastic effect would lead, in a general configuration subjected to constant displacement rate, to a form functionally similar to eq. (12), with perhaps a different constant of proportionality due to a different configuration of the flaw or multiaxiality of the loading.

As will be described later, a biaxial strip containing an initial crack of length $2c_0$ was therefore selected, loaded to failure at several different constant displacement rates, and the strain at failure was suitably multiplied by the characteristic material property. It was found that the γ_c so deduced was not constant, but did indeed vary with the failure time. Furthermore, tests were also repeated at several temperatures and it was found that the γ_c values so deduced not only varied also with temperature, but moreover all the time- and temperature-dependent data could be correlated to a typical WLF temperature-reduced time dependency, $\gamma_c = \gamma_c(t/a_T)$.

While it is obvious that the basic premise of a time-temperature-dependent cohesive energy density has not been completely established, the indirect proof and internal consistency of the data are believed to be convincing.

EXPERIMENTAL PROCEDURE AND RESULTS

The material tested was a butadiene–acrylonitrile–acrylic acid terpolymer crosslinked with an epoxy curing agent. From the mechanical standpoint, it exhibited rubbery behavior above 50°F in a constant strain test and viscoelastic behavior between 50° and −108°F. Equipment capability limited linearity tests to strains less than 18%, but the material obeys Boltzmann superposition to 18% strain as determined by stress relaxation measurements at several strains (Fig. 1). In addition, it was found that the moduli measured in uniaxial and biaxial stress states were interconvertible using the usual formulas of linear viscoelastic theory.[10]

The width of the strip specimen was $2w = 6.00$ in. Its height was $2b = 1.68$ in., thickness was 0.375 in., and it was bonded to strips of wood of
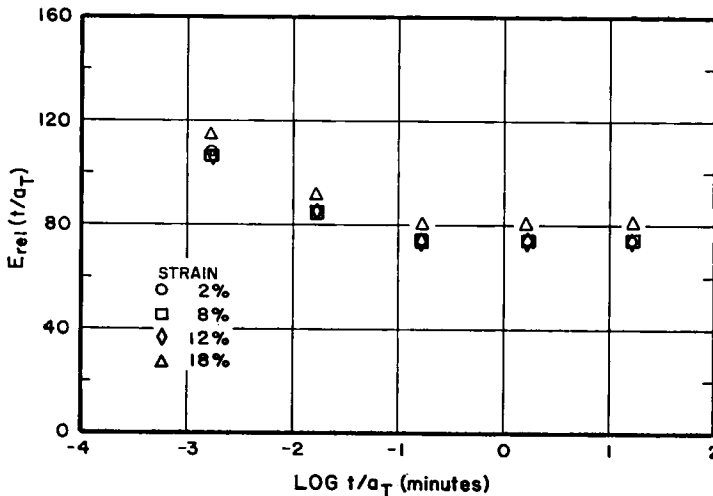
Fig. 1. Linearity of the relaxation modulus $E_{rel}(t)$. The glass modulus E_g was independently measured as 26,000 psi. Percentages shown are engineering strain.
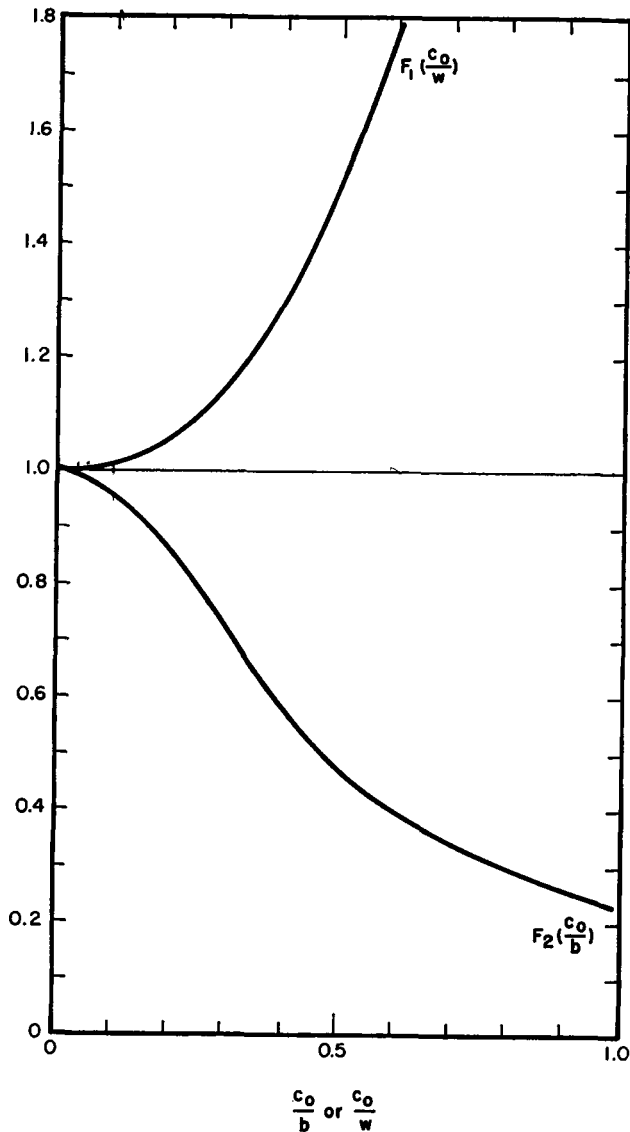
Fig. 2. Aspect ratio correction factors.

nearly the same thickness as the test specimen by a butt end joint. A precise line fracture 1.5 in. long was cut in the center of the specimen ($2c_0 = 1.50$). The wooden tab ends were held rigidly in a test assembly and displaced at a constant rate in an Instron testing machine. Elongation of the initial fracture was observed by two technicians, each making a mark on the load–time trace; the crack was also periodically photographed. A plot was then made of the crack length measured from the photos versus the time when the photo was taken. The resulting curve was extrapolated to

the intersection of the initial crack length. This point agreed well with the time of fracture initiation, t_0, observed by the technician. The rate of displacement, the load cell reading, and the test temperature were recorded, from which the average stress and displacement in the direction of loading could be calculated.

A stress analysis of the experimental geometry is available. Moreover, the Griffith analysis, which applies to a small crack in an infinite medium, can be corrected for the effect of the finite width and finite height of the actual specimen by a factor $F(c_0/w, c_0/b)$. Thus one can calculate the specific cohesive elastic energy in the configuration as

$$\gamma_c/c_0 = \left[\frac{\pi}{2(1 - \nu^2)^2} F\left(\frac{c_0}{w}, \frac{c_0}{b}\right)\right] E\ (v_0/b)^2 \qquad (14)$$

in which $v_0/b = \epsilon_y$ is the average axial strain imposed on the specimen. Actually, for the test geometry and the small crack being used, it was considered sufficient to correct for these two effects separately rather than simultaneously because the correction factor for their combined interaction amounted essentially to a second-order effect. Hence a Westergaard fac-
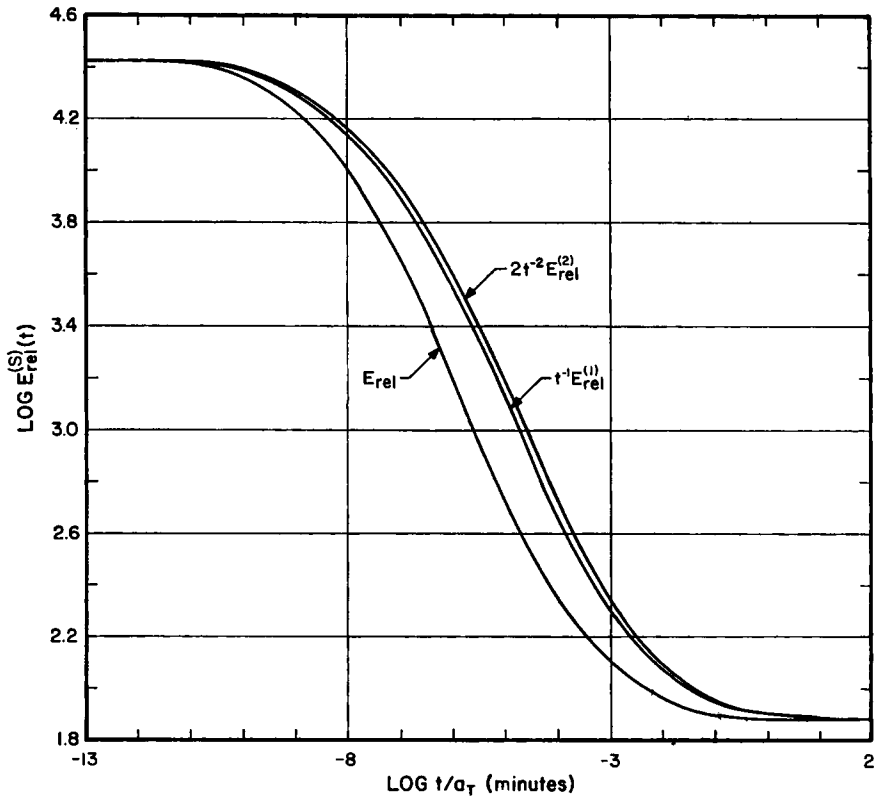

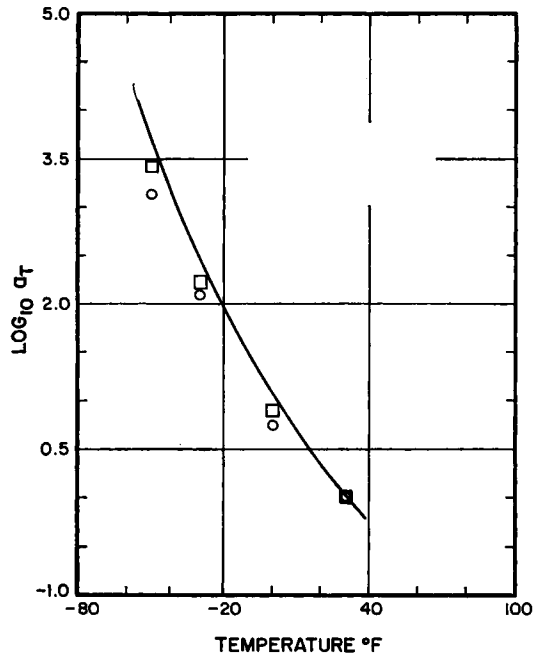Fig. 3. Relaxation modulus and its first two integrals.

Fig. 4. Comparison of empirical shift factors with WLF equation: (□) relaxation, modulus; (O) fracture energy; (—) WLF equation.
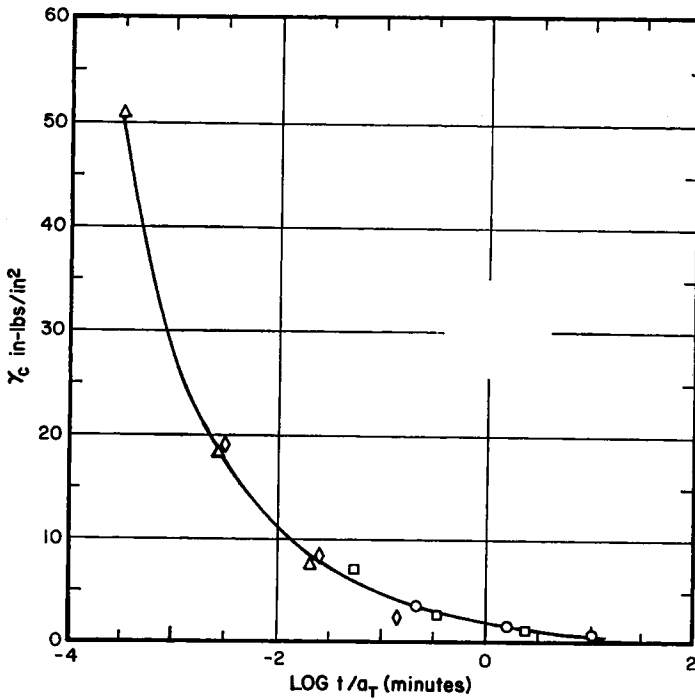

Fig. 5. Fracture energy: (O) 30°F; (□) 0°F; (◇) −30°F; (△) −50°F.

TABLE I. Experimental Data

Temperature, °F	Crosshead speed, in./min	Critical applied stress,[a] psi	Critical displacement, in.	Critical time, min	log t	log a_T	log t/a_T	$2t^{-2}E_{rel}{}^{(2)}$	γ_e, in.-lb/in.^{2}
30	2	22	0.22	0.22	-0.66	0	-0.66	86.0	3.59
	0.2	16	0.16	1.60	0.20		0.20	78.5	1.73
	0.02	11	0.105	10.5	1.02		1.02	77.0	0.73
0	2	30	0.295	0.295	-0.53	+0.74	-1.27	96.6	7.25
	0.2	14	0.19	1.90	0.28		0.46	84.1	2.61
	0.02	12	0.135	13.5	1.13		0.39	78.5	1.23
-30	2.	62	0.37	0.37	-0.43	+2.09	-2.52	162	19.1
	0.2	43	0.30	3.00	0.48		-1.61	106	8.22
	0.02	19	0.175	17.50	1.24		-0.85	88	2.32
-50	2.	119	0.425	0.425	-0.37	+3.14	-3.51	327	50.8
	0.2	72	0.355	3.55	0.55		-2.59	168	18.2
	0.02	37	0.275	27.50	1.44		-1.70	110	7.2

[a] Information data.

tor,[11] $F_1(c_0/w)$, was used for finite width and a Knauss factor[12] as corrected by Rice,[13] called $F_2(c_0/b)$, was used for the height, i.e., $F(c_0/w, c_0/b) \approx F_1(c_0/w)F_2(c_0/b)$. The separate correction factors, which depend on the aspect ratio of the specimen, are given in Figure 2. E is the elastic tensile modulus.

Note next that this functional form is essentially similar to that of the viscoelastic case,[12] viz.,

$$\gamma_c/a_0 = [(2t_0^{-2})E_{rel}^{(2)}(t_0)] \; \epsilon_\theta^2(a_0, t_0) \tag{15}$$

in which the equivalent viscoelastic modulus $[2t_0^{-2}E_{rel}^{(2)}(t_0)]$ replaces the elastic value E. Hence, assuming the spherical theory gives us the correct time dependence for the viscoelastic extension, we assume the viscoelastic equation for the specimen with a crack rather than spherical flaw is

$$\gamma_c/c_0 = \left[\frac{\pi/2}{(1 - \nu^2)^2} F_1\left(\frac{c_0}{w}\right) F_2\left(\frac{c_0}{b}\right) \right] [2t_0^{-2}E_{rel}^{(2)}(t_0)]\epsilon_y^2. \tag{16}$$

The relaxation modulus for our material was measured in actual relaxation tests on uniaxial bars (rather than being calculated from constant strain rate data) and is presented as a function of reduced time t/a_T in Figure 3 along with its normalized first two integrals. (It may be noted that there is not a very large difference between the successive normalized multiple integrals of the modulus, which justifies to some extent the use of merely $E_{rel}(t)$ as an approximate ad hoc engineering extension of the elastic fracture formulas to viscoelastic media, regardless of the load history.) The temperature shift factor a_T is shown in Figure 4.

The experimental data summarized in Table I, in which each data point is the average of at least three tests, has been reduced using eq. (16) in order to calculate $\gamma_c(t_0)$. These data are shown on Figure 5. The same WLF shift factor (a_T) as deduced for the relaxation modulus is seen to shift the cohesive energy data into a smooth curve. The Tobolsky formula[14] was used, in which a measured glass transition temperature, $T_g = -108°F$, was determined from thermal expansion measurement:

$$\log_{10}a_T = -17.44[(T - T_g)/(51.6 + T - T_g)]. \tag{17}$$

Figure 5 in conjunction with the shift factor therefore gives the time-temperature dependence of the specific cohesive energy density.

DISCUSSION

There are a few comments to be emphasized. First, one should recognize the shortcomings in an analysis wherein a cylindrical flaw model of a crack has been incorporated. It would be an improvement to obtain similar results using an actual crack configuration. So far, however, analytical difficulties have been encountered. Second, independent plane stress experiments have shown that different initial crack lengths lead to essentially the same time dependence in the cohesive energy, providing the crack is

somewhat longer than the thickness of the specimen.[15]   Third, the kinetics of the process during cohesive failure are presumably the same as during deformation, because the time-temperature shift factor is the same for $\gamma_c(t/a_T)$ as for $E_{rel}(t/a_T)$.   Finally, a similar investigation for the case of a filled material would be interesting because here the additional variable of adhesive debonding between the binder and filler is involved; presumably a different behavior of $\gamma_c(t/a_T)$ would result.

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