

Determination of Fracture Surface Energies by the Cleavage Technique

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Determination of Fracture Surface Energies by the Cleavage Technique

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The fracture surface energy of a material can be obtained by the cleavage technique, in which a crack is propagated along the median plane of a strip sample by forces applied at the free ends. Unfortunately, the stress distribution in a conventional sample is such that the crack tends to deviate from its original direction, so that, for isotropic materials, external constraints must be imposed to overcome this tendency. To interpret the data, relations have been derived, based on the assumption that simple beam theory can be applied to the system; this assumption is not strictly valid. The use of the constraints can be avoided by machining fine slots along the opposing faces of the sample, and the system can then be analyzed by a direct and unambiguous method. The modified technique has been applied to the glassy polymers poly(methyl methacrylate) and polystyrene for which comparative data are available from both tensile and cleavage experiments.

ACCORDING to the Griffith fracture theory,^{1,2} the amount of energy required to produce unit area of fracture surface, by extension of an existing defect, is an important parameter, which, inter alia, determines the ultimate properties of a material. The fracture surface energy (γ), defined in this way, may be regarded as a measure of the resistance of the material to the initiation of the fracture process. It can be obtained, using the analysis given by Griffith, from tensile experiments on samples containing defects of known size. The dependence of the ultimate tensile stress (T) on the size of the defect (c) is, according to the theory

$$T = K(E\gamma/c)^{1/2},$$

where E is the Young's modulus of the material and K is a dimensionless geometrical factor.

The results of such tensile experiments on samples of poly(methyl methacrylate) are illustrated in Fig. 1. The agreement of the data with the theoretical line is reasonably satisfactory, though there is, of course, a considerable degree of scatter in the results. The nature of the system is such that each sample gives only one data point, and hence a large number of experiments are necessary to establish the position of the theoretical line with any degree of assurance. Furthermore, application of the theory yields a value of the composite material constant $E\gamma$, and any uncertainty in the value of the elastic modulus increases that in the value of the fracture surface energy. Thus, although the tensile technique is experimentally straightforward, it is prodigal of time and material, and the uncertainty in the value of the fracture surface energy obtained is usually not less than about $\pm 30\%$. The uncertainty is small compared with the large discrepancy between the experimental and theoretical values of this parameter, but it is clearly much greater than is desirable if fracture surface energies of different polymeric materials are to be compared.

The criterion which Griffith adopted to define the condition of mechanical instability is

$$-\partial U/\partial c = \partial S/\partial c,$$

¹ A. A. Griffith, *Phil. Trans. Roy. Soc. London* **A221**, 163 (1921).

² A. A. Griffith, *Proc. Intern. Congr. Appl. Mech. (Delft)*, **55** (1924).

where U and S are the elastic strain and surface energies of the system, respectively. This criterion can be applied to systems other than that which Griffith himself considered, and some variants of the tensile case have been examined.^{3,4} Another which has received considerable attention is that in which a crack is propagated along the median plane of a strip sample, by applying forces at the free ends⁵⁻¹⁰ (Fig. 2). This is usually referred to as the cleavage technique. It is of particular interest from an experimental point of view, since it offers the possibility of obtaining the value of the fracture surface energy from a few experiments on a single sample. However, the techniques which have been used hitherto suffer from both theoretical and experimental shortcomings when they are applied to isotropic materials, such as polymers in the glassy state. It is the purpose of this article to discuss these shortcomings and to propose means whereby they may be overcome.

The details of the cleavage experiments described by different authors vary somewhat, and this leads to slight differences in the analysis of the systems, though the same principles are involved. To apply the Griffith instability criterion, it is necessary to establish the dependence of the elastic strain energy of the system on the length of the crack which it contains. The approach which has usually been adopted is to assume that the sample is symmetrical about the median plane and is equivalent to a pair of opposed identical cantilever beams, the lengths of which are given by that of the central crack. The strain energy of the system is then calculated from simple beam theory. However, the equations which have been applied to the problem are those appropriate to a cantilever beam which is built into a perfectly rigid support. This is equivalent to assuming that elastic energy is stored only in that

³ R. A. Sack, *Proc. Phys. Soc. (London)* **58**, 729 (1946).

⁴ H. A. Elliott, *Proc. Phys. Soc. (London)* **59**, 208 (1947).

⁵ J. W. Obreimoff, *Proc. Roy. Soc. (London)* **A127**, 290 (1930).

⁶ A. I. Bailey, *Proc. 2nd Intern. Congr. of Surface Activity* **3**, 406 (1957).

⁷ J. J. Benbow and F. C. Roesler, *Proc. Phys. Soc. (London)* **B70**, 201 (1957).

⁸ J. J. Gilman, *J. Appl. Phys.* **31**, 2208 (1960).

⁹ N. L. Svensson, *Proc. Phys. Soc. (London)* **77**, 876 (1961).

¹⁰ J. J. Benbow, *Proc. Phys. Soc. (London)* **78**, 970 (1961).

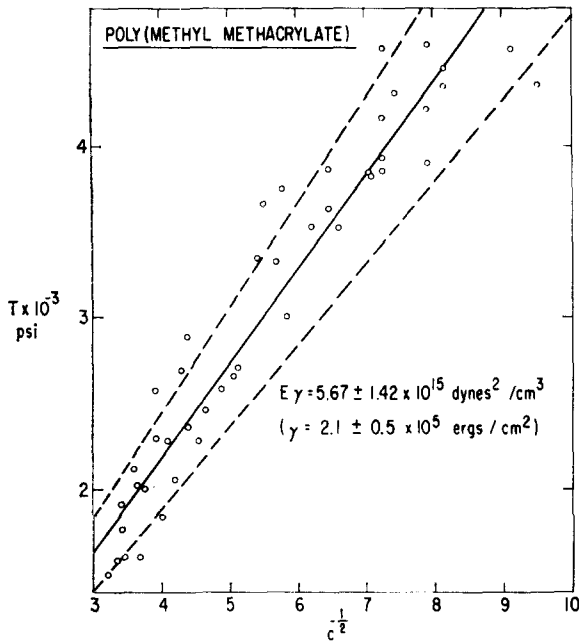


FIG. 1. Dependence of the tensile strength of poly(methyl methacrylate) samples on crack size.

part of a cleavage sample which is bounded by the surface of the central crack (A, Fig. 2). Clearly the (as yet) uncracked region (B, Fig. 2) will also be deformed, since it is not perfectly rigid as the simple beam theory demands, but has the same elastic modulus as the deflected region. Consequently the equations which have been derived for the elastic strain energy will be incorrect, for two reasons:

(1) Because of the deformation which it sustains, elastic strain energy will be stored in the uncracked region of the sample (B, Fig. 2). The extent of the deformation and, hence, the amount of energy stored, will depend on the local stresses at the root of the crack, which, in turn, depend on its length. Because of this dependence, a term corresponding to this stored elastic energy should be included in the equation defining the instability condition.

(2) Since the uncracked part of the sample is not perfectly rigid, the equations used to describe the characteristics of the deflected region (A, Fig. 2), including the stored elastic energy, will be inexact.

The influence of these factors is most apparent when the crack length becomes comparable to that of the sample. The deflection of the 'beams' is then obviously much less than the theory would predict from their length, while the uncracked region sustains extensive deformation. Under these conditions, the observed deviations from theoretical behavior, in particular the instability of the crack at fixed deflection of the sample, are not surprising.

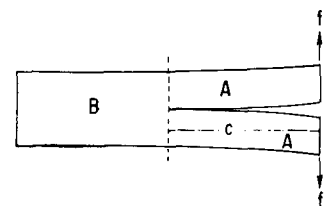
In practice, the two factors compensate each other to some extent since including the first increases the

total elastic strain energy of the system, while the corrections involved in the second reduce the calculated amount of energy stored in the deflected region. Nonetheless, the expressions which have been given for the dependence of the elastic strain energy on crack length cannot be regarded as wholly satisfactory.

In the two-dimensional model subjected to tensile forces, considered by Griffith,¹ the elastic strain energy function is obtained from the stress distribution equations. Unfortunately, the derivation of the corresponding equations for the cleavage sample is a problem of considerable complexity and has not been carried out. The correct analytical expression for the dependence of the elastic strain energy on the length of the crack in the deformed cleavage sample is likewise unknown.

A photoelastic study has been made of the stress distribution in the vicinity of the root of the crack in a cleavage sample. It indicated the nature of the deformation sustained by the uncracked region,¹¹ but did not explore the stress distribution over the complete sample as a function of crack length. However, the results relate to an experimental difficulty which tends to complicate the cleavage technique when applied to isotropic materials. The gradient of the maximum tensile stress in the vicinity of the crack tip is such that the crack tends to deviate from its median position and turn towards the edge of the sample,¹¹ a tendency which increases with increasing deviation. In isotropic materials, therefore, it is necessary to apply external constraints to the sample, to so change the stress distribution that the crack remains in the median plane.^{7,9,10} Unfortunately, the apparatus necessary to apply the constraints is clumsy, more or less, consisting of clamps or springs, and because of their effect on the system, it is usually impossible to determine the critical force necessary to engender instability. The critical deflection is measured, and beam theory is then used to analyze the data. Because of the considerations discussed above, this procedure cannot be regarded as wholly satisfactory. Only in the case of anisotropic materials (e.g., crystals) with specific cleavage planes, can the use of external constraints be avoided, but even here the simple cleavage technique can be applied only to particular planes in the crystal.⁸ However, a very simple modification in the configuration of the sample renders the use of constraints unnecessary, even for isotropic materials. Furthermore the mechanics of

FIG. 2. Conventional cleavage sample.



¹¹ R. Guernsey and J. J. Gilman, Proc. Soc. Exptl. Stress Anal. 18, (2), 50 (1961).

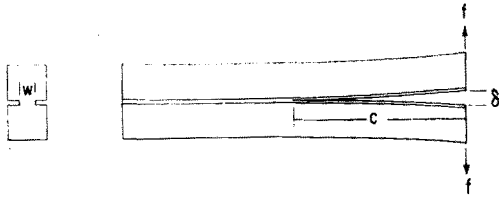


FIG. 3. Modified cleavage sample.

the system can be treated in a way which avoids the shortcomings of the analytical procedures noted above.

In the modified sample, the thickness in the median plane is reduced by machining fine slots along each face, so that the cross section is then as illustrated in Fig. 3. When a sufficiently large deflection is applied to the extremities of the sample, the crack increases in length while remaining in the region defined by the slots, and shows no tendency to deviate from this region. Since no external constraints are now needed to cause the crack to follow a precisely defined course, measurements of both force and deflection can be made.

THEORETICAL ANALYSIS

In analyzing the results, it is assumed that the relation between the length of the crack in the sample (c), the force applied to its extremities (f), and the total deflection which results (δ) can be represented by the equation

$$f = (ac^{-n})\delta. \quad (1)$$

This is a generalized form of the expression, derived from beam theory, which is normally used, viz.,

$$f = \left(\frac{3}{2}EIc^{-3}\right)\delta,$$

where I is the moment of inertia of cross section of one half of the sample and E is the elastic modulus of the material of the sample. According to both equations, the applied force is directly proportional to the deflection of the sample at constant crack length, and the slope of the force-deflection relation (m) varies inversely as some power of the crack length. From the assumed empirical expression

$$\log m = \log a - n \log c. \quad (2)$$

This equation can be used to check the validity of the empirical relation, and also to determine the value of the exponent n , which, according to beam theory, should be 3.

The elastic strain energy in the sample is given by

$$U = \frac{1}{2}f\delta = \frac{1}{2}a\delta^2c^{-n}. \quad (3)$$

Then

$$\left(\frac{\partial U}{\partial c}\right)_\delta = -na\delta^2/(2c^{n+1}) = -nf\delta/(2c). \quad (4)$$

If w is the width of the (reduced) cross section, at the fracture plane,

$$\partial S/\partial c = 2\gamma w. \quad (5)$$

Applying the Griffith criterion yields

$$(f\delta/w) = (4\gamma/n)c. \quad (6)$$

In this equation, f , δ , w , and c are all measurable quantities and the exponent n is easily obtained from the experimental data as indicated above. The equation predicts that the parameter $(f\delta/w)$ should be directly proportional to the crack length, the proportionality constant involving only the fracture surface energy, and the exponent n . In contrast to the tensile method, a knowledge of the elastic modulus is not required to evaluate the fracture surface energy, and thus one source of uncertainty in its value is eliminated.

Since the analysis of the system is based only on the assumption of an empirical equation, the validity of which can be determined experimentally, any slight deviations from the ideal form of the cleavage sample, such as nonplanarity of the crack, or dissymmetry of the sample, are no longer important.

EXPERIMENTAL

The fracture properties of the glassy polymers poly(methyl methacrylate) and polystyrene have been the subject of extensive examination by both tensile and cleavage techniques. Because of the availability of comparative data, it was decided to use these materials for the present study. The cleavage samples were cut from standard sheet stock; convenient sample dimensions were $11 \times 1\frac{1}{2} \times \frac{1}{4}$ in. The slots in the opposing faces of the sample were cut with a small circular saw, with a blade 0.006 in. thick. The height of the blade above the table and, hence, the depth of the slot which it produced, could be accurately adjusted. The depth of the slot was made the same in both faces, and to ensure stability of the crack, it was found that the slot should penetrate a distance of at least a quarter of the thickness from each face. The holes to take the loading pins were drilled $\frac{1}{4}$ in. from the end, and along the midline of each half of the sample.

An initial slot was cut through the sample, coplanar with the existing slots, to a distance equivalent to

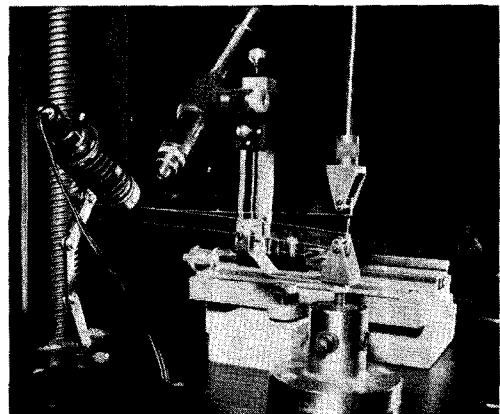


FIG. 4. Experimental arrangement for studying cleavage.

about one and a half times the total width, from the drilled end. Using the same saw which had been employed to machine the slots in the faces, it was found convenient to leave the end of the central slot with a "swallow-tail" configuration. A datum line was scribed on the face of the sample to define the point of load application, i.e., the position of the loading pins. The sample was then mounted in a conventional tensile testing machine, as illustrated in Fig. 4. A simple adjustable bracket was used to support the sample in a horizontal position. A cathetometer with a horizontal traverse was set up to observe the location of the crack tip, the direction of the traverse being parallel to the plane defined by the slots in the sample. The telescope was raised above this plane, however, and tilted so that its optical axis made an angle of about 45° to the horizontal, being focused, through the face of the sample, on the plane. A lamp mounted in the corresponding position on the opposite side of the sample directed a beam of light onto the crack, where it was reflected into the telescope. This arrangement rendered the tip of the crack readily visible. The cathetometer was zeroed on the point of suspension of the sample.

In starting the experiment, a natural crack was produced by increasing the deflection of the arms of the sample. Because of the "swallow tail" configuration of the end of the slot, the initiation of crack growth was usually quite smooth and easy to control. By manual adjustment of the extension controls of the testing machine, the crack tip could be brought to a convenient position, as observed through the telescope.

The most important feature of the cleavage sample is that under conditions of constant deflection, the system is inherently stable.¹² In practice this means that, if a deflection is applied to the system, sufficiently large to cause the central crack to increase in size, and this deflection is then maintained constant, the crack will grow only until it has attained an equilibrium length, determined by the values of the experimental parameters which satisfy Eq. (6). The values of the parameters having been obtained for this condition, the deflection is adjusted to some new, larger value, and equilibrium again established. The crack can thus be made to traverse the sample in a controlled manner, and a large number of data points can be obtained on a single sample.

A convenient experimental procedure is to apply a preselected deflection, and to determine the value of the force and crack length corresponding to it. After applying the deflection, the crack length increases and the measured force decreases and both approach their equilibrium values. Unfortunately, in such viscoelastic materials as the glassy polymers being studied, the force will also decrease under conditions of constant crack length and deflection because of relaxation effects, and hence the value measured in the type of experi-

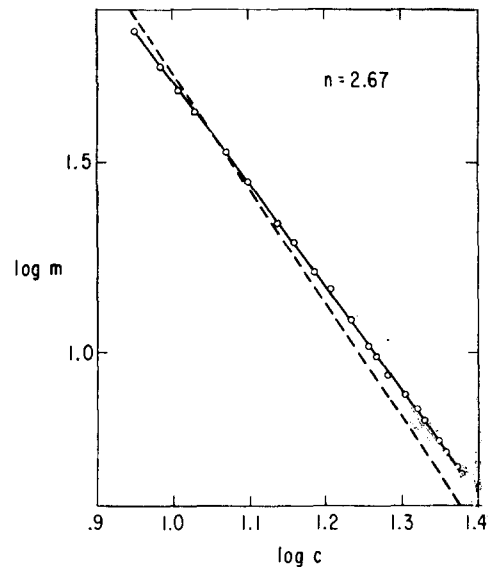


FIG. 5. Dependence of the slope of the force-deflection relation in cleavage on crack length.

ment described will be lower than would correspond to the true surface energy. To correct for these effects a cycling procedure has been adopted. It is assumed that on the initial deflection the sample is behaving essentially elastically provided that the extension rate is sufficient to attain the selected deflection in a reasonably short time interval (< 60 sec). This assumption is supported by the fact that the force-deflection plot is strictly linear. The deflection is maintained for a standard period (usually 10 min) during which both the force and crack length change in value, the decrease in the former being greater than would correspond to the increase in the latter if the sample behaved ideally. The sample is then brought to an unstrained condition, and allowed to remain in that state for as long as it was held under strain. The object of this procedure is to allow the effect of the relaxation processes to decay to a negligible value. When the same deflection is again applied, the maximum force recorded is greater than that observed at the end of the previous period of deflection, even though the crack length is the same. It is assumed that this force represents the true "elastic" value. However, because the applied force is now greater, the crack velocity increases, and it is again necessary to maintain the deflection to enable the system to approach equilibrium. The cycle is therefore repeated until, after the standard deflection is applied, the crack velocity falls to some arbitrarily low value ($\sim 10^{-4}$ cm/sec). This is usually attained after four deflections. The initial value of the force realized in the final deflection is then taken to be the equilibrium value corresponding to the measured crack length.

According to the assumed empirical equation, the relation between the slope of the force-deflection plot (m), and the crack length (c), should be given by Eq. (2). Good agreement with this relation is found over a

¹² J. P. Berry, *J. Mech. Phys. Solids* 8, 207 (1960).

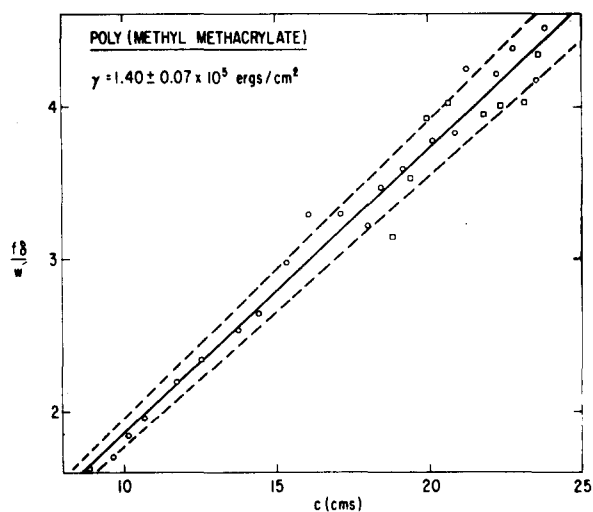


FIG. 6. Results of cleavage experiments on poly(methyl methacrylate). The points of different shape refer to experiments carried out on different samples.

considerable range of crack lengths (Fig. 5), though deviation occurs, because of end effects, for both short and long cracks. For this reason, measurements were confined to the central portion of the sample, at crack lengths greater than $2d$, and less than $(L-d)$, where d is the total width of the sample and L is its length. Normally only the data obtained under 'equilibrium' conditions are used to determine the value of the exponent n , but the same linear relation gives good agreement with the results from all deflections of the sample. Mechanical models of the system have also been studied. These were identical, in all dimensions, to the cleavage samples, but the central slot was produced by machining, rather than by extension of a crack, and, for each value of the slot length, only a single deflection was made. Again the results were in good agreement with those obtained in the cleavage experiment proper. The assumptions underlying the method of correction for viscoelastic effects thus appear to be well founded.

From the separated halves of the cleavage sample, the flexural rigidity (EI) can be obtained by a simple three-point bending test, and hence the beam theory equation appropriate to the system can be obtained explicitly. This equation is represented as the broken line in Fig. 5. It is clear from this figure that neither the slope nor the intercept of the best line through the experimental points agree with those expected from beam theory.

When the experiment is completed and the sample separated into two parts, the width of the fracture plane (w) is determined with the traveling microscope, at distances along the sample corresponding to the positions which the crack tip occupied when the "equilibrium" force-deflection measurements were made. From these results the function $(f\delta/w)$ is calculated and plotted against the crack length c . The data ob-

TABLE I. Fracture surface energies of glassy polymers ($10^{-5}\gamma$) ergs/cm².

Method	P.M.M.A. ^a	P.S. ^b	Author
Cleavage	4.9 ± 0.5	25.5 ± 3	Benbow and Roesler ⁷
Cleavage	4.5	9.0	Svensson ⁹
Cleavage	1.40 ± 0.07	7.13 ± 0.36	Present work
Tension	2.1 ± 0.5	17 ± 6	Berry ^{13,14}

^a P.M.M.A. = poly(methyl methacrylate).

^b P.S. = polystyrene.

tained on samples of poly(methyl methacrylate) are illustrated in Fig. 6, and those on polystyrene samples in Fig. 7. The agreement with the theoretical linear dependence is quite satisfactory, and the spread of the experimental points, as indicated by the broken lines, represents an uncertainty in the value of the fracture surface energy of $\pm 5\%$. In these figures the points of different shape refer to the results of experiments on different samples. This degree of precision is much better than can be obtained by the tensile technique and, as indicated in Figs. 6 and 7, a satisfactory value of the surface energy can be obtained on a single sample, and in a much shorter time than is possible by that technique.

RESULTS

The results of the present experiments are summarized in Table I, where a comparison is made with the results obtained by other workers^{7,9} using similar techniques, and with the results of experiments on tensile samples.^{13,14}

DISCUSSION

It is clear, in performing these experiments, that there is a certain degree of arbitrariness involved in defining the "equilibrium" condition from which the

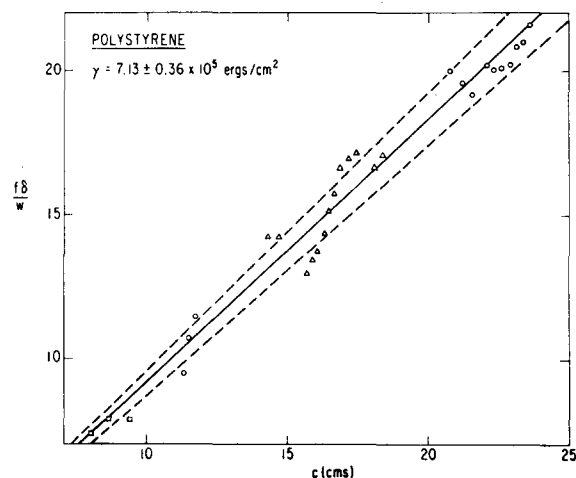


FIG. 7. Results of cleavage experiments on polystyrene. The points of different shape refer to experiments carried out on different samples.

¹³ J. P. Berry, *J. Polymer Sci.* **50**, 107 (1961).

¹⁴ J. P. Berry, *J. Polymer Sci.* **50**:313 (1961).

surface energy results are calculated. A true steady state, at constant deflection, could not be realized, and the crack continued to increase in size at a slow but measurable rate when the experiment, at any particular deflection, was terminated. It seems probable that in polymeric materials a true equilibrium condition may never be realized, and that the crack will continue to grow so long as the sample is in a strained condition. This is also indicated by observations which have been made on the growth of crazing cracks.¹⁵

The time dependent factors discussed above make it somewhat difficult to compare the values of fracture surface energy obtained by different techniques and by different workers. In cleavage experiments carried out at constant deflection, the values obtained will depend on the time scale of the experiment, both because of relaxation effects and because of the persistence of crack growth. Benbow and Roesler⁷ considered that the crack was stationary if its velocity was less than 10^{-3} cm/sec. They also state that an experiment on one sample, which presumably involved a number of determinations at different crack lengths, took about 15 min. Svensson⁹ determined the "equilibrium" length of the crack 5 min after adjusting to a selected deflection. It seems probable that the experimental time scales, and the terminal fracture velocities, were comparable in the work described by these authors. The results obtained in experiments on poly(methyl methacrylate) are also comparable, and are higher than those found in the present experiments, in which the time scale was much longer and the terminal crack velocity correspondingly less ($\sim 10^{-4}$ cm/sec) (Table I). Also, in the present experiments corrections were made for relaxation effects by the application of the cycling procedure discussed above. No such corrections were made by previous workers.

The results of the experiments on polystyrene samples show a much greater variability (Table I), a factor which is probably due to the more erratic behavior of this polymer. The morphology of the crack is usually complex, and there is a tendency for the nature of the crack propagation to change suddenly from a slow to a fast mode.^{7,9,10} It appears from the work of Benbow that these effects are related to the molecular weight of the material,¹⁰ and it is possible that the differences in the results reported may have arisen from this cause. However, the results of the present cleavage experiments and those from experiments in tension¹⁴ were obtained on the same material.

In the experiments described above, the procedure is such that the crack is constrained to move and the end point is taken when the moving crack comes to a halt. However, in tensile experiments the fracture surface energy is derived from the conditions which obtain when a stationary crack begins to increase in size.

From theoretical considerations it would be expected that the former condition would give a lower value of the surface energy than is obtained when the crack is initially at rest.¹² From experiments carried out on the same material, the expected trend in the results is obtained (Table I).

It is possible in cleavage experiments to define the instability condition in terms of the force required to cause the stationary crack to grow in size, and this technique has been used successfully for mica⁶ and for crystals.⁸ In the present study some attempts were made to follow this procedure, but the results were very erratic and showed much greater variability than was found in the method finally adopted. A slow approach to the critical condition is necessary to avoid exceeding it; but the longer the time taken to achieve the condition, the more serious become the effects of relaxation. No satisfactory compromise for these opposing tendencies could be found.

EXTENSION OF THE MODIFIED CLEAVAGE TECHNIQUE TO OTHER MATERIALS

Some crystalline materials, with specific cleavage planes, can be cleaved successfully without the use of external constraints or reduction in cross section at the site of the fracture plane. However, the data from experiments on such materials could be better interpreted by the use of an analysis based on a verifiable empirical relation, as discussed above, rather than on one related to the demonstrably inapplicable beam theory. In such crystals, only certain particular lattice planes are normally susceptible to cleavage. The experimental technique described in this paper raises the possibility of determining the energy of cleavage of other planes to which the conventional methods cannot be applied.

In the materials which have been investigated in this study, the crack length can be observed directly, and a very simple analysis of the data is possible. The experimental method described can also be applied to opaque materials, but in this case some modification of the analysis of the system is required since the length of the crack is unknown.

From Eq. (1) and (6), the critical (equilibrium) condition is given by

$$f^{(n+1)} = (4w\gamma/n)^n a \delta^{(1-n)}. \quad (7)$$

Hence,

$$\log f = \frac{n}{(n+1)} \log \left(\frac{4w\gamma}{n} \right) + \frac{1}{(n+1)} \log a - \left(\frac{n-1}{n+1} \right) \log \delta. \quad (8)$$

Thus there should be a linear relation between the logarithm of the equilibrium force and the logarithm of the applied deflection, the intercept being a function of the fracture surface energy γ . However, the expres-

¹⁵ J. A. Sauer and C. C. Hsiao, Trans. Am. Soc. Mech. Engrs. 75, 895 (1953).

sion for the intercept also includes the constant a . Both this constant and the exponent n can be obtained from force-deflection experiments on a model cleavage sample, as described above, in which the central slot is produced by machining rather than by extension of a crack.

CONCLUSION

The fracture surface energy is, potentially, a useful index of the ultimate properties of a material. The

cleavage technique, as described above, provides a method of determining this parameter with reasonable accuracy, and with the expenditure of a limited amount of time and material.

ACKNOWLEDGMENT

The author wishes to express his thanks to Peter F. Erhardt for his assistance in carrying out much of the experimental work described herein.

Rhe-Optical Properties of Polymers. III. Dynamic Polarized Light Scattering

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In a recent paper on the dynamic strain-optical coefficient for polyethylene, it was reported that the strain-optical coefficient displayed a relaxation-dispersion effect. Experiments have now been carried out to measure the dynamic scattering strain-optical coefficient as a function of frequency. Preliminary results are presented and interpreted in terms of the motion of domains within the specimen under dynamic conditions.

MEASUREMENTS of the mechanical properties of polymers yield information concerning their moduli and loss factors, as a function of time, temperature, and frequency. However, much more information can be generated by observing also the effect of mechanical deformation on the optical properties.^{1,2} In the case of semicrystalline polymers, the relationship between the mechanical and optical properties and the reorganization of internal structure is less clear. This is mainly a result of the increased complexity of the material involved. For example, in strained polyethylene under static conditions, x-ray diffraction, light scattering, birefringence, and infrared dichroism serve as tools to indicate macromolecular as well as micro-molecular reorganizational changes, which although similar in behavior have not as yet yielded to a unified theory. More recently, studies of the dynamic strain-optical and scattering strain-optical coefficients have indicated that much more information can be generated, which should aid in the development of a generalized theory of the molecular and macroscopic time and temperature dependent mechanical properties of polymers.

Recently we have carried out experiments using polarized light-scattering techniques and present here some qualitative results of our study of the polarized dynamic scattering strain-optical coefficient of low-density polyethylene.

¹ D. G. LeGrand and P. F. Erhardt, *Trans. Rheol. Soc.* **6** (to be published).

² D. G. LeGrand and P. F. Erhardt, *J. Polymer Sci.* (to be published).

THEORY

The use of light scattering to study the structure of polymers has been exploited by many investigators.³⁻⁵ The scattering of light from inhomogeneous bodies arises as a result of the fluctuations in optical properties within the system. From studies of light scattering from polymer films, information concerning the size, refractive-index differences, anisotropy, and orientation of domains can be obtained.

Stein⁶ has shown that the intensity of light scattered by a polymer film containing both orientation and density fluctuations for the I_{V_v} component will be given by

$$I_{V_v} = K \left\{ \langle \eta^2 \rangle_{av} \int \gamma(r) \frac{\sinh r}{hr} r^2 dr + \frac{4}{45} \langle \delta^2 \rangle_{av} \right. \\ \left. \times \int f(r) \left[\frac{\langle \eta^2 \rangle_{av}}{\alpha^2} \gamma(r) + 1 \right] \frac{\sinh r}{hr} r^2 dr \right\}, \quad (1)$$

where $\gamma(r)$ represents a correlation function for fluctuations in average polarizability defined by

$$\gamma(r) = \langle \eta_1 \eta_2 \rangle r / \langle \eta^2 \rangle_{av}, \quad (2)$$

³ F. P. Price, "Investigation of Crystal Growth in Polyethylene by Light Scattering," in *Growth and Perfection of Crystals*, edited by R. H. Doremus, B. W. Roberts, and D. Turnbull (John Wiley & Sons, Inc., New York, 1958).

⁴ J. J. Keane and R. S. Stein, *J. Polymer Sci.* **20**, 327 (1958).

⁵ R. S. Stein and J. J. Keane, *J. Polymer Sci.* **17**, 21 (1958).

⁶ R. S. Stein, J. J. Keane, F. H. Norris, F. A. Bettelheim, and P. R. Wilson, *Ann. N. Y. Acad. Sci.* **83**, 37 (1959).