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Stress Wave-Initiated Fracture in Amorphous Thermoplastics

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Synopsis

Several brittle thermoplastic materials, including polystyrene, SAN, and PMMA, were tested in tension using a spectrum of testing rates up to 15,000 in./min (630 cm/sec). At test velocities below 7000 in./min (294 cm/sec), the materials behaved "classically" with strength properties increasing, elongation decreasing, and single fractures generally forming. At rates between 7000 and 9000 in./min (294 and 378 cm/sec), a transition from classical to stress wave-initiated fracture was observed. The mechanical properties, the nature of the fracture, and the nature of the crazing preceding fracture were shown to be profoundly affected when the materials were strained at rates sufficient to initiate stress wave fracture. Under these conditions, the apparent load-carrying capabilities of the materials were reduced by an order of magnitude.

INTRODUCTION

Modern applications of polymers have begun to more fully tax the chemical and mechanical capabilities of this class of materials. For example, automobile flexible front-end structures must resist the severe environment posed by prolonged exposure to sunlight and atmospheric agents as well as sustain the large stresses and deformations encountered when a 4000-lb (1815-kg) mass is arrested from 5 mph (8 kilometers/hr) without damage. Just as sunlight and atmospheric agents are recognized as severe environments, so should rapidly applied large loads be recognized. Resistance to this new set of factors should be designed into the polymers beginning with their chemical structure and continuing to final part design.

The mechanical properties of polymers have been studied, in most cases, at relatively slow loading rates. The ASTM standard test rate for tensile testing, for example, is 0.2 in./min (8.4×10^{-3} cm/sec). Under these conditions, the applied load is more or less uniformly distributed throughout the sample with the load as measured representing the resistance of the molecules and the molecular interactions to deformation. Initially, at small strains, the response is elastic, molecular coils are extended but still retain the ability to return to their original configuration. At larger strains, broader, more permanent deformations are encountered which are not fully recoverable upon unloading. As the process is continued, the means by which additional straining is accommodated are consumed. At some value of strain, a critical value of stress is achieved and the specimen fractures. The effect of increasing test rates has been described in great detail, particularly for materials above their glass transition points (T_g), by Ferry[1] and Tobolsky.[2] In general, a Boltzmann-type superposition is found relating increasing test rate with lowering test temperature. The expected response to increased test rate is, then, increasing strength factors; i.e., modulus, accompanied by decreasing elongation at break.

A somewhat different set of circumstances leads to the eventual fracture event in materials at temperatures below their T_g. In this glassy state, the concepts of fracture mechanics are applicable[3]; and the measured average stress level again reflects the resistance of molecules and interactions to deformation. Berry[4] has shown that the flaw theory as developed by Griffith is valid in the description of the fracture process in glassy polymers. Such flaw theories require that a uniform distribution of microscopic flaws exists in all materials. When external loads are applied elastic deformation begins with the flaws acting as sites of stress concentration. As straining continues, a critical value of stress is achieved at a flaw. Fracture is then initiated and propagates, providing that sufficient energy from elastic strain potential energy or external sources is present to generate the new surface area and to accomplish the plastic deformation known to occur at the crack tip. It would appear that after the crack initiation stage, the crack tip is the most effective flaw in the system, and single fractures are expected and generally occur in low rate testing. It is also known that surface flaws are often the fracture initiation sites.

With the exception of the voluminous impact data of the Izod and Charpy type, relatively little systematic study[5] of the effect of loading rate on glassy thermoplastics has been carried out. It could be expected from the theories of Ferry and Tobolsky that the elastic modulus should increase and the extent of plastic deformation decrease with increasing test rate. The work of Ely[5] confirms this expectation.

The present work was initiated to study a number of thermoplastic materials below their T_g under a spectrum of test rates. Such results will allow a better understanding of the mechanical response of these materials in applications where rapidly applied loads can be expected.

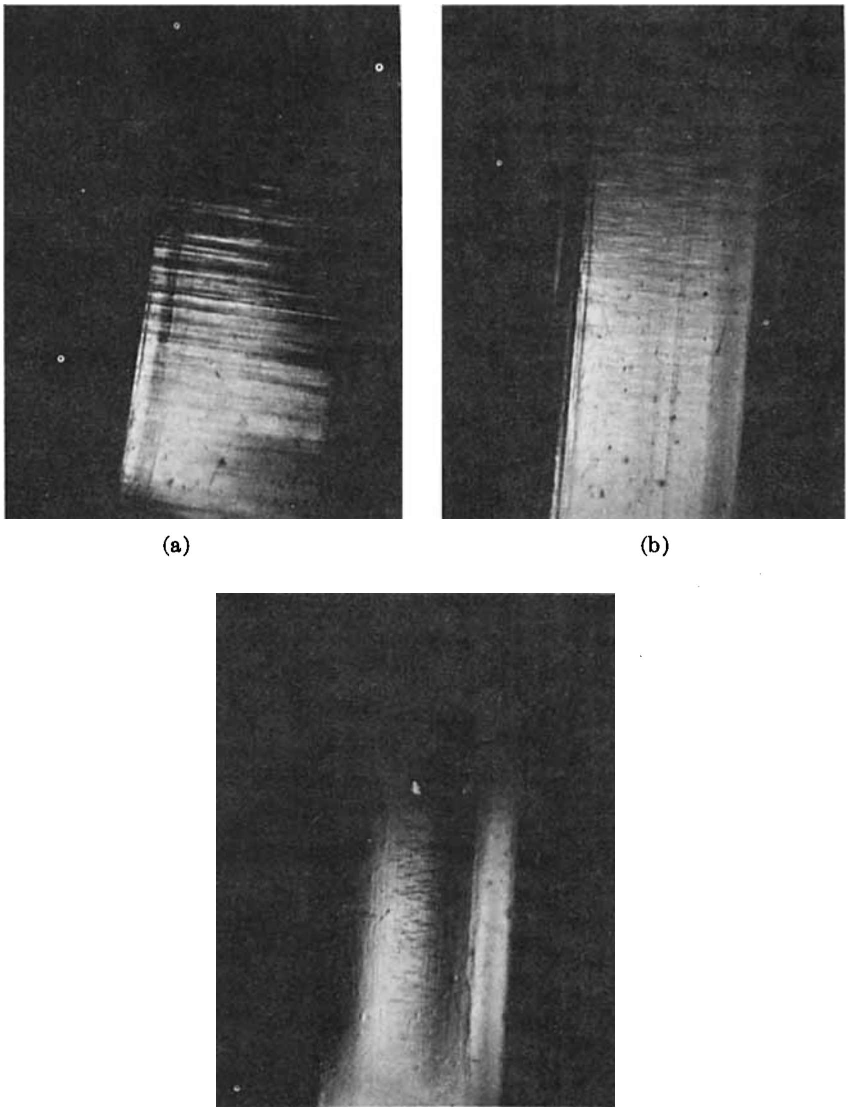
EXPERIMENTAL

Materials used in this study were general-purpose injection-molding grades of polystyrene (Shell 300), poly(styrene-co-acrylonitrile) (Dow SAN 760), and poly(methyl methacrylate) (Rohm and Haas PMMA 100). Sample test bars were prepared by injection molding, under conditions recommended by the material suppliers. The tensile bars were 2.5 in. (6.35 cm) in total length with a 0.625 in. (1.59 cm) reduced section length, 0.150 in. (0.38 cm) section width, and a thickness of approximately 0.125 in. (0.317 cm). The radius of curvature of the approaches to the reduced section of the test bar was $^5/_8$ in. (1.59 cm). At least six samples of each material were tested at each of five test rates. Average values are given along with the 95% confidence limits of the distribution of results. Test rates generally used were 150, 3000, 6500, 10,000, and 15,000 in./min (6.3, 126, 273, 420, and 630 cm/sec). This contrasts with the earlier work of Ely which terminated in the 3000 to 5000 in./min (126 to 210 cm/sec) range. Tensile testing was carried out on a Plastechon Model 581 tester under room temperature conditions. The tester is driven by rapidly applying nitrogen or helium gas at 200–300 psi (1.4–2.1 MPa) to the top of a piston attached to the moving portion of the test machine. At the end of the driven piston is a hydraulic reservoir with the test speed being controlled by allowing the hydraulic fluid to be forced from the reservoir through an adjustable valve. The load transducer, a Bytrex type PL 1000, is suspended above the driven ram by a very massive solid structure. Since a finite time is required to accelerate the ram to a given speed, a slack adapter was introduced to allow for this acceleration period before the test specimen was contacted. Because of the rigid interconnection of the driven assembly and the load-measuring transducer along with the relatively massive clamping jaws, the phenomenon of ringing occurs at test speeds above 3000 in./min (126 cm/sec) with materials of the type used in this study. This ringing manifests itself as a decaying sinusoidal perturbation of the load cell output. The procedure which has come to be accepted in handling this phenomenon is to average out the artificial perturbation. This was done where applicable in this study. After the initial acceleration of the ram, the deformation and fracture of the specimen removes energy from the system, resulting in decreasing ram velocity. The test rates listed are, therefore, smoothed average values of the test speed only during the deformation and fracture of the specimen. To gain further insight into the deformation and fracture processes as they occur, certain critical tensile tests were photographed using a Hycam high-speed camera with the capability of producing 4×10^4 pictures per second. Electron photomicrographs of the more important fracture surfaces were also prepared.

RESULTS AND DISCUSSION

As outlined in the Introduction, it was expected that we should observe increasing strength properties, decreasing elongations with single fracture initiation sites as we progressed toward higher test rates. Elongations as measured by a linear variable differential transformer (LVDT) attached to the moving ram of the test machine showed, essentially, the expected result up to a test velocity of around 7000 in./min (294 cm/sec). Tensile strength with somewhat wide 95% confidence limits due to extensive ringing also showed essentially the expected response up to the same test velocity. Below this test velocity, single fractures were generally encountered with the various materials. Polystyrene in particular showed extensive crazing with the entire reduced section of the test bars completely filled with fine reflective craze surfaces progressing to just short of the surface of the bars. The SAN bars showed less evidence of crazing, while PMMA showed virtually no crazing. Figure 1 shows a comparison of the degree of crazing associated with low-rate, 150 in./min (6.3 cm/sec), tensile testing of the three materials.

As test velocities were increased to the 9000 to 10,000 in./min (378 to 420 cm/sec) range, the nature of the material responses changed completely. The first most apparent change was the initiation of numerous fracture sites. In general, the reduced section of the test bars virtually exploded into small pieces. The recovered pieces were found to be cubical in nature and of the order of $^1/_{16}$ to $^1/_4$ in. (0.159 to 0.635 cm) in thickness. The tensile (ultimate) strength which had shown evidence of a gentle increase significantly decreased a full order of magnitude in the case of polystyrene. Crazing, which had been wide spread, particularly in polystyrene, was drastically reduced and was confined to fracture



(a) (b) (c)

Fig. 1. Craze patterns. Test speed 150 in./min (6.3 cm/sec). (a) Polystyrene, (b) Poly(styrene-co-acrylonitrile) (SAN), (c) Poly(methyl methacrylate) (PMMA).

sites. It appeared that some type of critical test velocity had been achieved and that the mechanism of the fracture process had been altered.

From the literature, the early work of Hopkinson[6] best paralleled our findings. In that study, iron wires were impulsively loaded in the axial direction by falling weights. The drop height (velocity) was increased until fracture occurred. When that drop height was maintained, it was found that the weight could be reduced, yet fracture would still occur. The work concluded that a critical velocity existed where the fracture process became independent of impacting energy or momentum and dependent only on velocity. The fracture stress was being provided by discrete stress waves traveling through the wire rather than by stresses generated by classical means, i.e., material resistance to deformation.

The subject of transient wave propagation in materials of varying sorts has been extensively studied, with polymers receiving relatively little attention. Kolsky[7] has presented the details of stress wave propagation in solids. This whole subject is very complex in nature with only the very simplest cases being tenable to analysis. A tensile bar of the type used in this study represents a very complex geometry. The test procedure of maintaining the loading rate throughout the test also adds to these already complex circumstances. The materials used in this study were in general considered "brittle," i.e., fracture occurred during the initial linear portion of the stress–strain curve. If we take the liberty to estimate the wave velocity and stress parameters of these viscoelastic materials with the appropriate elastic counterparts and consider only the longitudinal wave for our qualitative description of the stress wave fracture phenomena, we may write that[8]

$$\text{wave velocity} = C_L = [3K(1 - v)/p(1 + v)]^{1/2} \qquad \text{(a)}$$

where K = bulk modulus, p = density, and v = Poissons ratio;

$$\text{instantaneous stress} = p\ C_L\ V \qquad \text{(b)}$$

where p = density, C_L = wave velocity, and V = impacting velocity.

The stress may be tensile or compressive in nature. These waves may be reflected or refracted from free surfaces and made more complex by the fact that a tension wave when reflected comes back as a compression wave. When two or more waves collide, both stress attenuation and/or intensification may occur. With the preceding representing a most limited discussion of the nature of stress waves in solids, we will describe the response of the materials of this study as we progressed from classical to stress wave initiated fracture.

TENSILE STRENGTH RESPONSE

The tensile responses of the test materials are shown in Figure 2. The low rate response is typical of "classical" fracture with breaking stress rising slowly with increasing test rate. In the range between 7000 to 9000 in./min (294 to 378 cm/sec), a transition from classical to stress wave fracture occurs. The conventional method of measuring stress, i.e., load supported by the whole bar, becomes inadequate, and an apparent order of magnitude loss in strength is observed. During this transition the impacting velocity term of eq. (b) becomes large enough that the stress level attendant to the wave surpasses the critical stress level for fracture. At this point, the fracture energy is provided by the highly localized and transient wave front and not from uniform resistance to deformation.

To pursue this somewhat further, a series of tests were run with the test velocity constant at 10,500 in./min (440 cm/sec) and varying temperatures up to 105°C. The resistance to deformation from a classical fracture view should decrease with increasing temperature, leading to reduced stress at a given elongation. The results of these tests are shown in Figure 2. The effect of the increased temperature was very minor and did not result in reversing through the stress wave fracture transition to the classical mode of fracture. The lack of

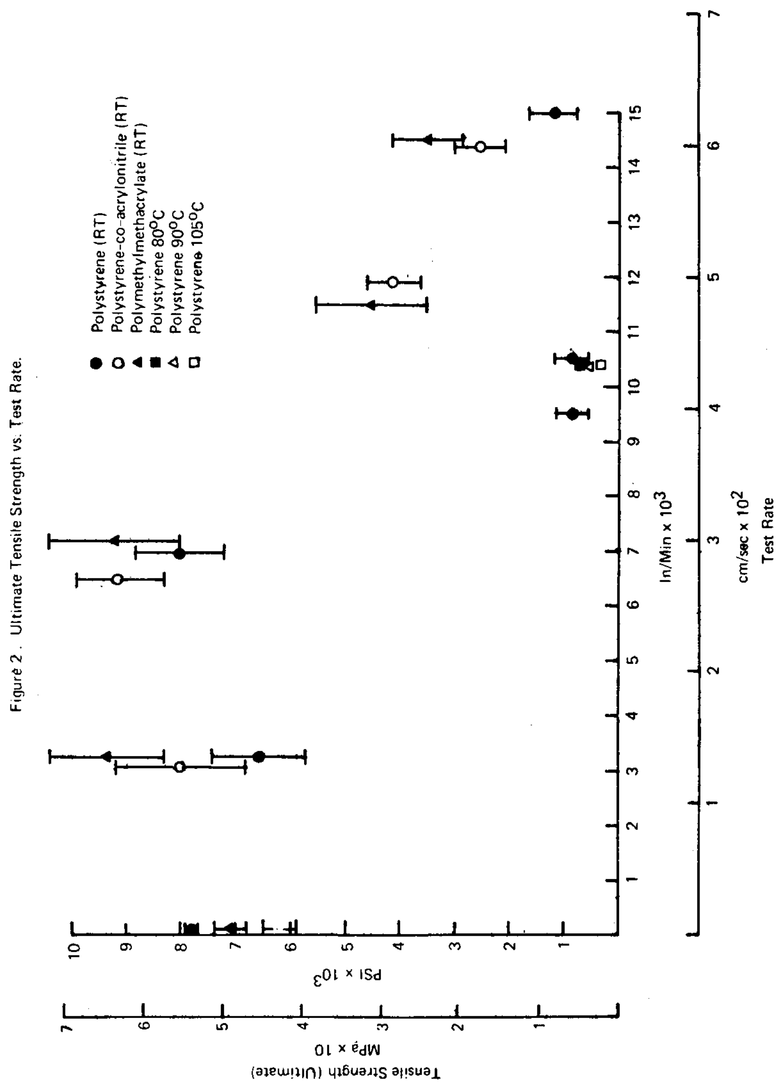


Fig. 2. Ultimate tensile strength vs. test rate.

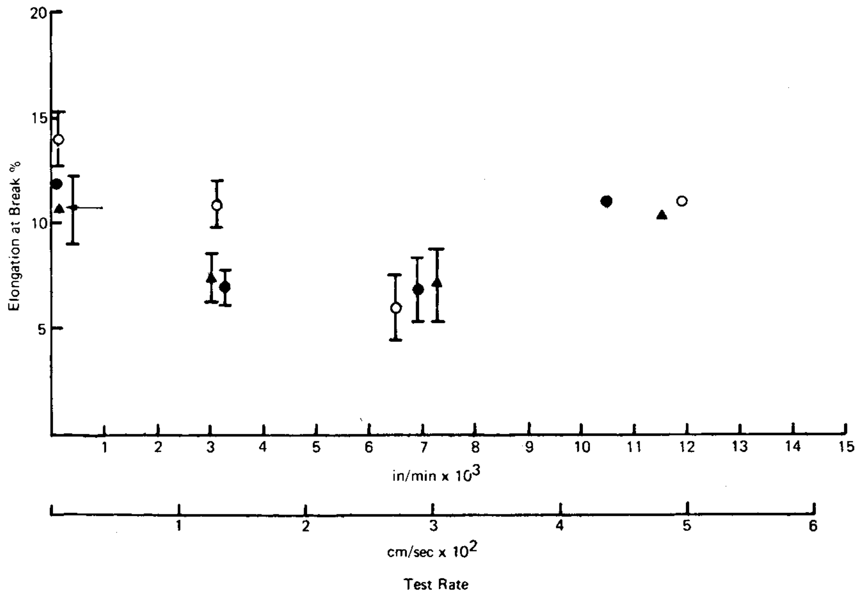


Fig. 3. Elongation at break vs. test rate. (●) Polystyrene, (○) Polystyrene-co-acrylonitrile, (▲) poly(methyl methacrylate).

temperature effect reemphasizes the fact that the energy for fracture originates from a wholly different set of circumstances in the stress wave case.

The elongation to fracture (Fig. 3) also behaved peculiarly as the classical-to-stress wave fracture transition was approached. At test speeds below 7000 in./min (294 cm/sec), the elongation was found to decrease with increasing test velocity as predictable from classical theory. In the stress wave fracture region, a reversal toward higher elongation was found. The data plotted in Figure 3 were obtained from three sources. For rates up to 20 in./min (0.83 cm/sec), an Instron machine was used. In the intermediate region up to 7000 in./min (294 cm/sec), the LVDT trace for ram displacement was correlated with the duration of the load cell output on Plastechon 581. Above this speed, elongations were obtained by analysis of high speed motion pictures of fracture events carried out on the Plastechon 581. The phenomenon of delayed fracture, perhaps due to an extended period of elastic deformation, has been previously noted in glassy polymers and in glass by Kolsky.[9]

EFFECT ON THE NATURE OF FRACTURE

As described in the Introduction, a consideration of the Griffith criteria of fracture leads to the conclusion that after a crack initiates, it should propagate to completion, yielding a single fracture through the specimen. This is particularly true under the conditions of the tests herein described where loading was continued throughout the test. Single fractures were, indeed, generally the case at test velocities below 7000 in./min (294 cm/sec). In the stress wave fracture region, however, fractures as shown in Figure 4 became the most common fracture mode. The figure shows the results of a test on an SAN specimen at a rate of 10,500 in./min (440 cm/sec). The view is through the 0.150 in. (0.38 cm) width

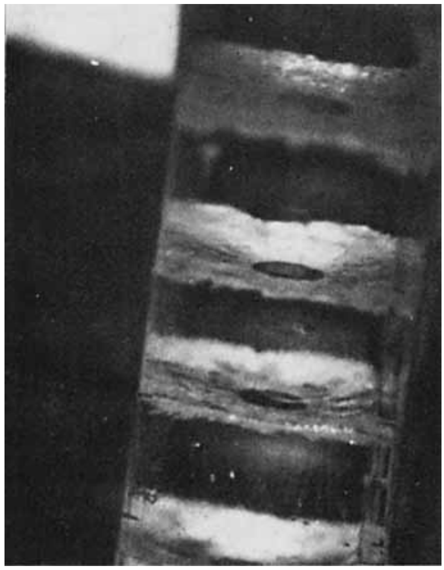


Fig. 4. Polystyrene-co-acrylonitrile fracture segment. Test speed 10,500 in./min (440 cm/sec).

of the reduced section of the test bar which was 0.125 in. (0.317 cm) thick. The bar segment was tilted at an angle of approximately 30° from horizontal to show the detail of the fracture faces. This bar was somewhat peculiar in that three of the fractures did not propagate completely through the surface layers of the specimen. The more common response was for the fractures to go to completion and reduce the specimen to a number of cubical segments. In this figure, four complete and evenly spaced fractures are seen. The fractures, initiated at the center of the bar, propagated slowly, leaving a mirror-like disk and finally propagated at a higher rate leaving the rougher surface. The nature of the fracture surfaces will be discussed in a later section of this paper. Between the second and third full fractures, an isolated disk section may be seen. This appears to be a fracture which initiated, proceeded through the slow growth stage, but did not advance to the rapid crack propagation stage. A very plausible explanation for the type of multiple fractures experienced in this work may be found in a work by Rinehart[6] describing the comminution of brittle rock-like materials. In that work, the very intensive stresses generated by the interaction of a stress wave with its own reflection from a free surface are used to describe the phenomenon of multiple spallation. The governing equation for the stress in this situation is of the form of eq. (b) (described previously); however, the velocity term is made substantially larger due to the interaction of the tensile and compressive wave components. The implication of this explanation is that the individual fractures as seen in Figure 4 must form successively, one after the other.

To pursue this further, high-speed motion pictures were made of an SAN fracture experiment run at 12,000 in./min (500 cm/sec). A Hycam camera was used at a framing rate of 28,000 pictures per second. Selected frames of the film are shown in Figure 5. The two dark lines and the circles were used to measure the total strain and the local lateral and axial strains as the test progressed. The time between successive frames was 3.6×10^{-5} sec. The fractures are numbered

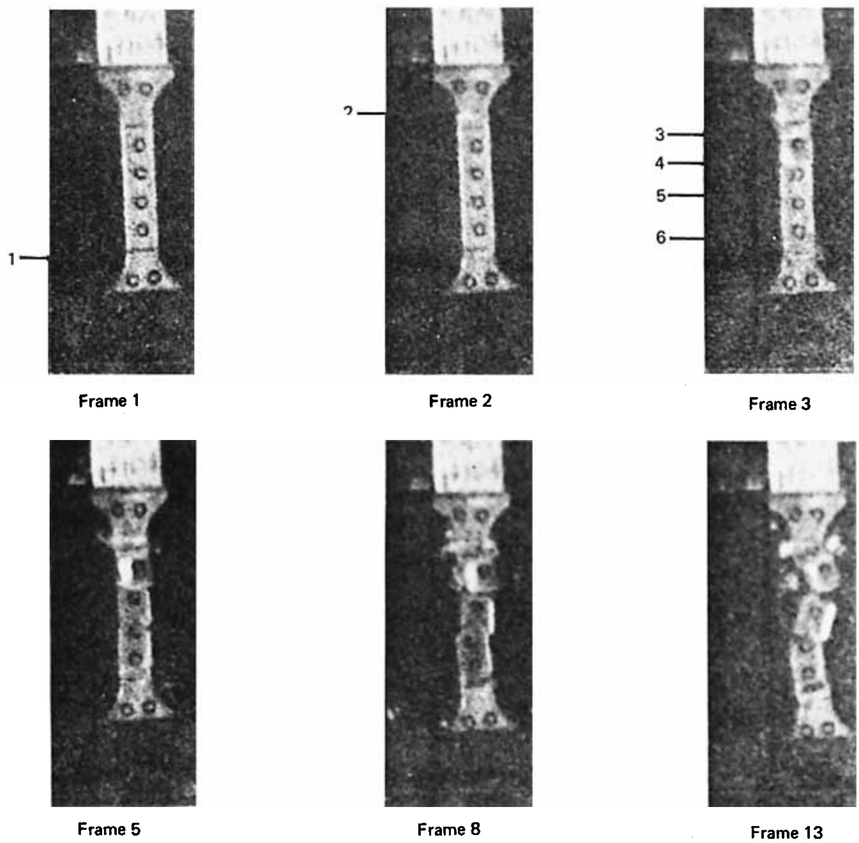


Fig. 5. High-speed photographs—poly(styrene–co–acrylonitrile) fracture. Test speed 12,000 in./min (500 cm/sec); camera speed 28,000 pictures/sec.

in order of appearance. The first fracture, although difficult to see in the photograph (Fig. 5), did not progress completely through the specimen until all of the other fractures had formed. In this test, six separate fractures occurred. The first and second cannot be explained by the multiple spallation phenomenon. The first fracture apparently initiated when the wave which originated in the lower (driven) jaw progressed into the reduced section of the test bar. The second fracture occurred when components of the wave reflected from the opposite end of the bar progressed into the other end of the reduced section of the bar.

This situation may be similar to the work described by Kolsky[10] in which waves were propagated through cones of PMMA. The conclusion of that work was that since the momentum of the wave must be conserved as it propagated through the specimen of decreasing cross section, the velocity (V) term as used in eq. (b) must be increased. In those tests, fractures were noted near the apex of the cone. The central four fractures (Fig. 5) do seem to be of the spallation type and appear to form successively.

In Rinehart's work,[8] it was stated that the shape (stress distribution) of the wave was an important aspect of the multiple fracturing. He also stated that a portion of the total energy and momentum of the wave was retained in the fractured segments. This generally resulted in the segments flying off at some velocity proportional to the amount of momentum they retained. Based on these findings, some conclusions as to the shape of the wave in this study may be deduced.

Figure 5 shows that the first segment, between fractures 2 and 3, virtually explodes beginning at frame 5. When the film strip from which Figure 5 was derived was viewed as a motion picture, the second segment was seen to rotate wildly as the remaining segments formed and fell away. All of this indicates a relatively broad stress distribution with a sharp peak in stress near the head of the wave. Kolsky[11] found highly stressed natural rubber to be dispersive in nature indicating that the larger amplitude (most highly stressed) components of the wave traveled faster than the smaller amplitude components, resulting in a sharp peak approaching shock wave intensity at the head of the wave. The materials in this study appear to respond in a similar manner.

EFFECT OF INCREASED RATES ON CRAZING

The phenomenon of crazing has been described in a variety of polymeric materials by Kambour[12,13] In general, the process appears to consist of the generation of voids followed by either the coalescence of these voids or the drawing of the bulk material from the void containing areas of the sample.[14] In this study, the polystyrene samples were seen to form the most profuse crazing and will be used as an example in a description of the effect of increased testing rate on the nature of crazing.

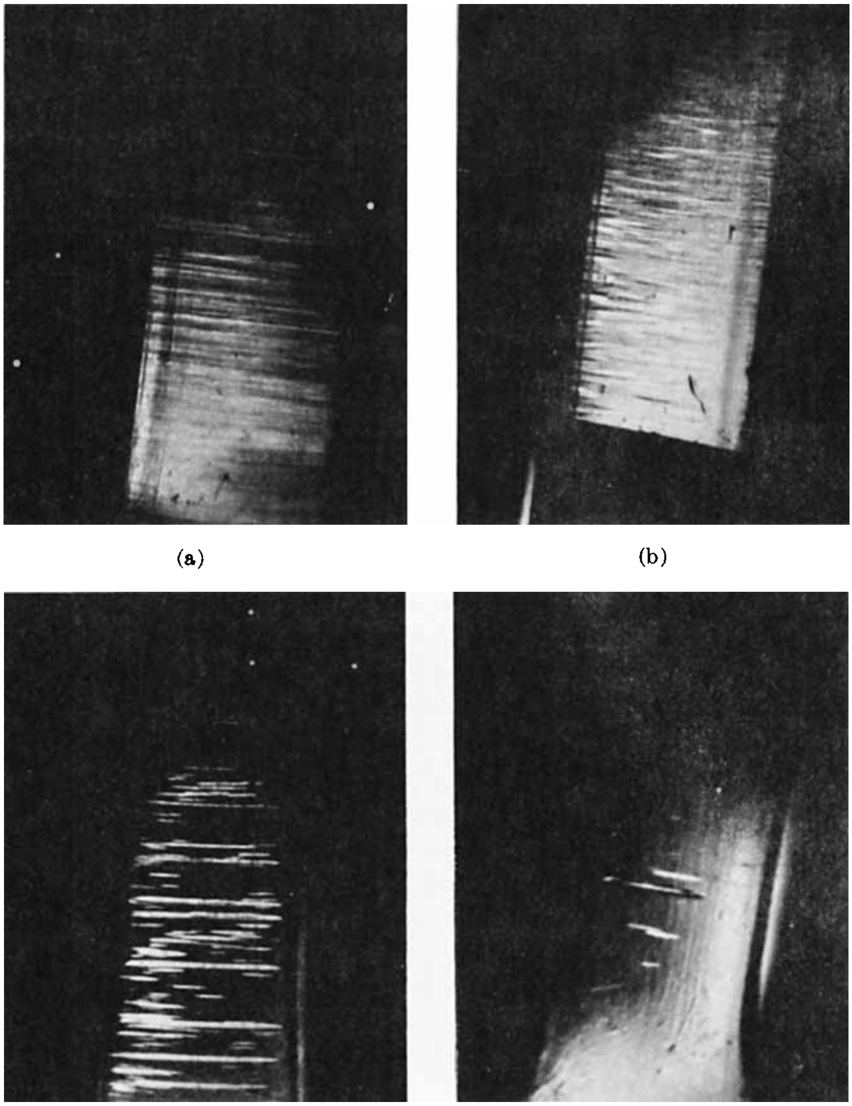
Figure 6 shows the extent of crazing obtained at rates varying from 100 to 10,500 in./min (4.2 to 440 cm/sec). When observed visually, the crazes formed at or below 100 in./min (4.2 cm/sec) are very profuse, and the bar is milky in appearance. From Figure 6 it is readily apparent that, as the rate is increased, fewer and fewer crazes are formed and the tendency toward shorter crazes predominates. In Figure 6d, a stress wave fracture, very few crazes were found. The largest feature in 6d, in fact, was seen to be a fully formed and fractured center disk.

Crazes are a form of localized ductile deformation and as such should obey a critical shear stress criteria at least for the growth step. As the test speed becomes higher, less time is available for the shear stresses to develop and less crazing is seen. In the stress wave case, the high tensile stresses are present for only a very short period of time at any one place, and crazing in general becomes attendant only to fracture.

FRACTURE SURFACE EFFECTS

The nature of the actual fracture event can, to a large degree, be deduced from an analysis of the fracture surfaces. In many brittle material fractures, however, the surfaces are very complex, and a tenable analysis is not possible. In this work, the SAN and PMMA surfaces were in general found to be of this complex type as seen in Figure 7. Polystyrene fractures provided substantially more information as to the fracture mode in these high rate tests.

Figure 8a shows a general view (50×) of a polystyrene sample fractured at a test rate of 10,500 in./min (440 cm/sec). The center disk is a very prominent feature of the micrograph, and close scrutiny of this region shows another more

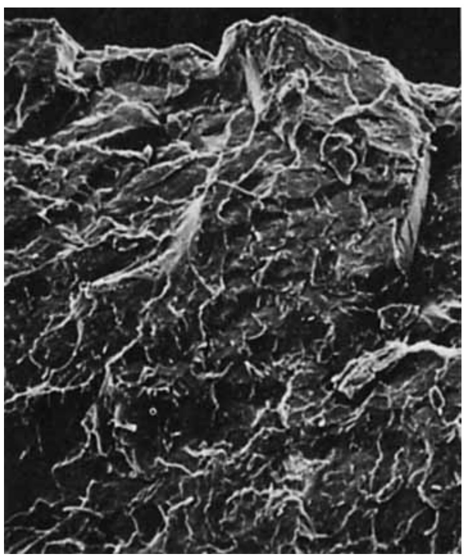


(a) (b) (c) (d)

Fig. 6. Polystyrene crazes at varying test speeds. (a) 100 in/min (4.2 cm/sec), (b) 3000 in/min (126 cm/sec), (c) 7000 in/min (293 cm/sec), (d) 10,500 in/min (440 cm/sec).

diffuse disk within the larger disk. A very sharp transition between the larger disk region and the rough regions may be seen. Figure 8b is a 3000× magnification of the disk area. The region is characterized by a very planar surface with secondary fracture sites randomly distributed throughout the area. A fine general texture is visible and probably is the broken and retracted craze fibrils. Figure 8c is the very complex, rough fracture area viewed at 3000× magnification. This region is very similar in appearance to the PMMA and SAN fracture in Figure 7. A higher magnification (10,000×) of this region is shown in Figure 8d. A fine general texture is again seen, indicating that crazing was a precursor to final fracture in the rough area as well as in the disk area.

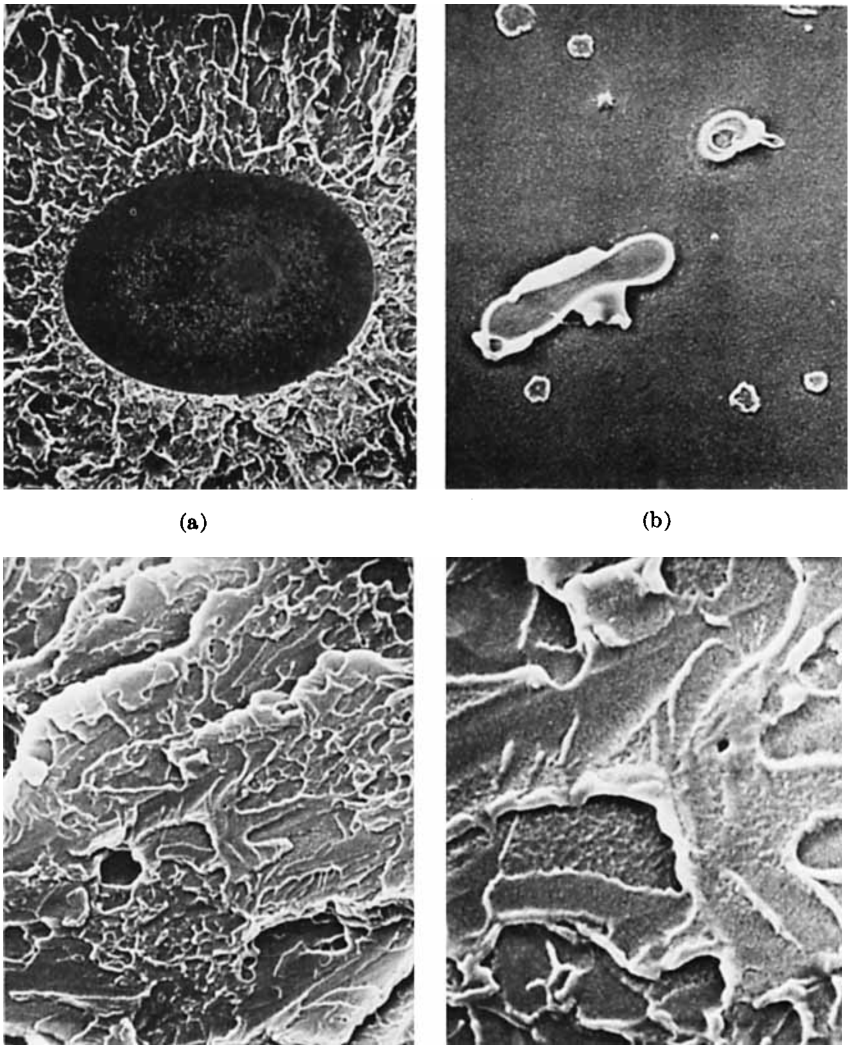
(a)



(b)

Fig. 7. Fracture surface micrographs 50×. Test speed 10,500 in./min (440 cm/sec). (a) Poly(styrene-co-acrylonitrile), (b) Poly(methyl methacrylate).

The appearance of these fracture surfaces may be best understood in light of the work carried out by Clark and Irwin.[15] In that work, the relationship between crack extension force and crack propagation velocity was developed. Conditions were defined based on the nature of the stress field attendant to the crack tip under which crack forking could be expected. Figure 8a then appears to show the crack initiation (diffuse disk) followed by crack acceleration (remainder of the large disk). Finally, a limiting crack velocity is reached, while the



(a) (b) (c) (d)

Fig. 8. Polystyrene fracture surface micrographs. Test speed 10,500 in./min (440 cm/sec). (a) General View 50X, (b) Disk Area 3000X, (c) Rough Area 3000X, (d) Rough Area 10,000X.

crack extensional forces continue to increase, resulting in multiple forking which gives rise to the rough surface appearance.

Samples tested at 100 in./min (4.2 cm/sec) did not show any evidence of the rough surface appearance, indicating that crack extensional velocity is a function of test speed. The analysis of the fracture surfaces showed definite differences among the materials used in this study. The polystyrene and SAN at rates below 7000 in./min (294 cm/sec) did not attain crack velocities which would give rise to the rough texture. PMMA, on the other hand, always produced the rough surface, making it impossible in most cases to distinguish the crack origination point (Fig. 7). This would indicate that the limiting crack velocity was attained almost instantly in the case of PMMA. SAN fractures above 7000 in./min (294 cm/sec) generally produced only the rough texture (Fig. 7) but did on occasion produce the smooth disk with the transition to the rough texture (Fig. 4). Polystyrene at high rates always produced surfaces as shown in Figure 8. All of this indicates that even at very high test rates in brittle, glassy materials, some molecular mobility is present and is governing the mechanical responses of these materials.

CONCLUSIONS

The mechanical properties of the polystyrene, SAN, and PMMA used in this study were shown to be profoundly affected when strained at rates sufficient to initiate stress wave fracture. The load-carrying capability under these circumstances could in some instances be reduced by a full order of magnitude. Significant changes were noted in the very nature of the fracture process as well as with the initiation and propagation of the craze structure preceding fracture. All of these effects were found to occur at test rates of around 7000 in./min (294 cm/sec).

The nature of stress waves is profoundly affected by geometry; and, therefore, it is difficult to assess the effect of such waves on complex molded articles. We have, however, shown the possibility of stress wave-initiated fracture at test velocities realized in many current polymer applications.

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