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The measurement and origin of fracture toughness in polyethylene

Strebel, Jeffrey Jerome, Ph.D.
Case Western Reserve University, 1993

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THE MEASUREMENT AND ORIGIN OF FRACTURE TOUGHNESS
IN POLYETHYLENE

by

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Submitted in partial fulfillment of the requirements
for the Degree of Doctor of Philosophy

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GRADUATE STUDIES

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THE MEASUREMENT AND ORIGIN OF FRACTURE TOUGHNESS IN POLYETHYLENE

Abstract

by

JEFFREY JEROME STREBEL

Fatigue crack propagation studies on annealed and quenched MDPE specimens showed the lifetime of annealed specimens to be one third that of the quenched specimens. The less tortuous crack path of the annealed specimens was traced to a voided and relatively weak annealed root craze, as compared to a continuous tough quenched root craze. DSC and DMS revealed a multiphase annealed morphology but a more homogeneous quenched morphology. The larger quantity of tie molecules was believed to be a major factor in the improved fracture resistance of the quenched specimens.

A method to calculate fracture toughness, $J_{1C}$, from fatigue tests was developed by utilizing the energy definition of the $J$ integral. In HDPE, it was shown that the new fatigue method yields $J_{1C}$ values which are nearly identical to those obtained using the standard ASTM bending method. Using the new method, specimen thicknesses, and hence morphologies, can be tested which more closely approximate the thickness of the real component. In addition, the new method permits the calculation of $J_{1C}$ from brittle fractures of some tough MDPE resins which do not demonstrate brittle fracture with the ASTM method.

By varying the loading conditions, different amounts of damage are
produced at the propagating fatigue crack tip. Since the J integral is a measure of the crack tip deformation field, $J_{1C}$ was observed to vary with loading conditions. A linear relationship between the length of the damage zone and $J_{1C}$ was observed to be independent of loading conditions, specimen geometry, and material.

Since brittle failures in polyethylene generally occur as a result of the material being exposed to low stresses for long periods of time, and since $J_{1C}$ was not able to distinguish differences in fracture resistance between different MDPEs, a method which utilizes time and energy to calculate the "power to fracture" is introduced. The proposed method yields the same "power band" for one MDPE tested under a variety of conditions and is able to distinguish between different MDPEs.

Fatigue crack propagation studies were conducted on two very different specimen geometries. Extensive damage analysis was conducted to determine the types and extent of crack tip damage which developed in both specimens. The summation of these damage types is proportional to the resistance moment, which was employed in the crack layer theory to calculate the specific enthalpy of damage and the dissipation coefficient. Both specimens had the same specific enthalpy of damage, $\sim 300$J/g, and the dissipation coefficients, $\beta_{SEN} = 4.6 \times 10^{-5}$, $\beta_{arc} = 1.1 \times 10^{-4}$, were inversely proportional to the number of cycles to failure.
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CHAPTER 1:
THE EFFECTS OF ANNEALING ON FATIGUE CRACK PROPAGATION IN PolyETHYLENE

ABSTRACT

Fatigue crack propagation tests on annealed and quenched medium density polyethylene showed the annealed specimens to have much lower resistance to crack initiation and subsequent propagation. Although the same fracture mechanism, in which the brittle crack gradually becomes more ductile, prevailed in both cases, the voided and fibrillated crack tip root craze in the annealed material was much weaker than the nonfibrillated quenched root craze. Microstructural analyses indicate that the annealed material was a multiphase system, while the quenched material had a more homogeneous morphology. The highest melting fraction of the annealed material was composed of lamellae that were 270Å thick, while the quenched lamellae averaged 160Å in thickness. The reduced fatigue crack propagation resistance of the annealed material was suggested to be a result of a lower concentration of tie molecules.
INTRODUCTION

Semicrystalline polymers are often envisioned as two phase systems in which crystallites are supported in an amorphous matrix. The morphological composition of semicrystalline polymers can be altered by variations in molecular structure or thermal history. For example, a linear polyethylene may be able to crystallize into thick well formed lamellae, while a branched polyethylene may have more crystal defects and thinner lamellae depending on its branch type and branch distribution. However, because there are many molecular parameters which are interlinked with each other and with the morphologies they could spawn, it is often difficult to firmly establish cause and effect relationships particularly in regards to long term fracture resistance. Even when such relationships could be determined, the results may be disputed by other studies. One such case shows that while some authors have suggested that crack propagation resistance improves with decreasing spherulite size [1,2], others claim spherulite size has no effect [3-5]. Increases and decreases in slow crack propagation resistance have been attributed to increasing crystallinity [4-7]. The molecular variables mentioned above alter the fatigue crack propagation rate through the type of morphology which they produce. In this work, the molecular variables have been held constant by restricting the study to one medium density ethylene copolymer. However, the morphology has been altered by changing the thermal history. Both thermal histories, the annealed and the quenched state, have a distinct fatigue crack propagation behavior. This paper attempts to correlate the different morphologies with the changes that they induce in the fatigue cracking behavior.
EXPERIMENTAL

Material

The medium density polyethylene employed for these studies was a gas pipe resin which was supplied by Osaka Gas Company. The resin had a weight average molecular weight of 123,000 g/mol, a molecular weight distribution of 9.7, and it contained 6.5 butyl branches per one thousand carbon atoms.

The resin was compression molded into four millimeter thick plaques. Some plaques were quenched in ice water, while others were allowed to slow cool in the press. The slow cooled plaques were then annealed at 120°C for 240 hours. Single edge notch (SEN) specimens were milled from the plaques; final specimen dimensions were 145 mm x 20 mm x 4 mm. The notch was introduced at mid-height by pressing a razor blade to a five millimeter depth.

Fatigue Crack Propagation Tests

Fatigue tests were conducted on an MTS servohydraulic machine at a frequency of 1 Hz. Three experiments for each thermal history were conducted to ensure reproducibility. Load control experiments were conducted using a sinusoidal loading wave form which varied from a maximum stress of 6.66 MPa to a minimum of 0 MPa. Maximum loads in this range (~25% σy) have previously been shown to produce brittle fatigue fracture of pipe resins in reasonable periods of time [8].

During the fatigue test load - displacement hysteresis loops were recorded on an analog x-y recorder. Crack tip observations and crack length measurements were made using a travelling optical microscope.

Scanning Electron Microscopy (SEM)

Fracture surfaces were gold coated and examined using a Jeol JSM-840A scanning electron microscope. Crack profile specimens were first
etched to remove microtome damaged material and elucidate the microscopic details. The etching procedure is a modification of that used by Olley, Hodge, and Bassett [9]. The specimens are stirred in a five percent KMnO₄/H₃PO₄ solution for two hours, the solution is then diluted with water to a fifth of the original concentration. After thirty minutes in the diluted solution, the specimens are rinsed in water and then soaked in thirty percent H₂O₂ for ten minutes. After the H₂O₂ soak, the specimens are rinsed in water and mounted on the SEM specimen holders. A small metal wedge is inserted in the crack opening to hold the crack open to its maximum crack opening displacement during the fatigue test. The specimens were then sputtered with palladium. This procedure facilitates viewing of the craze microstructure.

Transmission Electron Microscopy (TEM)

Polyethylene morphology was examined using a single stage replication technique [10]. The specimen to be examined was first etched with KMnO₄/H₃PO₄, as described in the SEM section. Platinum/Carbon was sputtered on to the etched surface at about a thirty degree angle in order to create contrast. Over this layer a continuous carbon coat is applied for physical integrity. A three percent solution of poly(acrylic acid) (Carbopol 907), PAA, in water is then placed on the carbon coat and allowed to dry. The dried PAA, carbon, carbon/Pt sandwich is then gently lifted form the surface with a forceps. The sandwich is floated on water to dissolve form the surface with a forceps. The sandwich is floated on water to dissolve the PAA. The remaining carbon and carbon/platinum layers are then lifted from the water on a copper microscope grid (200 mesh). The replicas were examined at 100 kV in a Jeol JEM-100SX.

Differential Scanning Calorimetry (DSC)
Differential scanning calorimetry was conducted using a Perkin Elmer 7 series DSC. The samples were heated from 30 to 180°C at 10°C/min. Cooling and reheating curves were recorded at the same rate. Sample weights were six to eight milligrams. The percent crystallinity was determined by dividing the heat of fusion by that of a perfect polyethylene crystal - 65.9 [11]. The Thomson equation [12-13] was employed to estimate lamellae thickness from the peak melting temperature.

Wide Angle X-Ray Diffraction (WAXD)

Wide angle x-ray diffraction was performed on a Philips XRG3100/APD3520 system. A continuous scan was recorded from 2 to 44° (2θ). The percent crystallinity was calculated by dividing the sum of the areas of the 110 and 200 peaks by the entire area, including the amorphous contribution, under the spectra in this region (16°-26°).

Dynamic Mechanical Spectroscopy (DMS)

Rectangular bars 3.1 x 10 x 43 mm were tested in shear on a Rheometrics RMS-800. A temperature/frequency sweep was performed in which the response of the specimen to a 0.05% strain was recorded. Data was recorded at four degree intervals from -150 to 125°C. The temperature was allowed to equilibrate for 1.5 min after each temperature change. During each interval, G', G", and tan δ was recorded at five different frequencies 10, 5, 1, 0.5, and 0.1 Hz.

RESULTS

Material Characterization

Before examining the fatigue cracking results, it may be best to examine the variable material morphologies induced by the two thermal treatments. Thus, in an effort to determine differences in crystallinity and lamellae
thickness, differential scanning calorimetry (DSC) was utilized. The percent crystallinity was determined by dividing the heat of fusion by the heat of fusion of a perfect polyethylene crystal - 65.9 cal/g [11]. The annealed state showed a crystallinity of 54.7%, while the quenched had a crystallinity of 49.0%. These figures were confirmed by wide angle x-ray diffraction (WAXD): annealed, 57.8%; quenched, 49.7%. The percent crystallinity by itself is insufficient to explain crack propagation resistance, since some have shown an increase in fatigue crack propagation resistance with increasing crystallinity [5-7], while this study and another [4] indicate decreasing fatigue crack propagation resistance with increased crystallinity.

The DSC traces show three separate peaks for the annealed and only one peak for the quenched (Fig 1). The separate annealed peaks are indicative of segregation on a supermolecular level. The low temperature melting peak and the low temperature shoulder are indicative of material that was excluded from the thickening lamellae (high temperature peak). This multiple peak melting behavior signifies a multiphase system. On the other hand, the quenched condition produces a single smooth peak; there is no segregation in the quenched state.

It is expected that the higher crystallinity and multiple peaks induced by annealing are a result of lamellar thickening. The lamellar thickness may be estimated from the Thomson equation [12-13],

$$L(\lambda) = \frac{2\sigma_e}{\Delta H_f} T_0 (T_0 - T),$$

where $T_0 = 414.5^\circ K$, $\sigma_e = 0.070 \text{ J/m}^2$, $\Delta H_f = 288 \times 10^6 \text{ J/m}^3$ [13]. Using the melting temperature of the largest peak, the approximate lamellae thickness for the annealed condition was calculated to be 70% greater than that of the quenched (Table 1). Obviously, this annealing treatment promotes lamellar
thickening.

Since the fatigue resistance of a polymer is related to its dissipative capabilities, the dynamic mechanical spectra (DMS) of both thermal histories were obtained (Fig 2). The dissipative capability, as measured by \( \tan \delta \), was higher for the quenched material at the temperature of the fatigue experiments (23°C). This increased ability to dissipate energy contributes to the greater fatigue resistance of the quenched state. Because of its greater crystallinity, the annealed condition showed a higher storage modulus \( (G') \) than did the quenched. The loss modulus \( (G'') \) of the annealed was also higher because its higher crystalline content permits more crystalline relaxations [14]. Geil showed similar trends in HDPE, however his annealing treatment did not effect changes from the quenched state for his copolymer pipe resins [10]. This lack of change may be a result of his more mild annealing condition (110°C, 12 hours) or because of molecular differences (ie. molecular weight, branch number) between his pipe resin and ours.

The dynamic mechanical spectra also confirm the DSC results. The annealed \( \tan \delta \) curve shows three crystalline transitions labeled \( \alpha_c, \alpha_c', \text{ and } \alpha_c'' \) which have relative intensities similar to the three transitions observed in DSC. The \( \alpha_c'' \) and \( \alpha_c' \) transitions occurred at about 55 and 105°C, respectively. The \( \alpha_c \) transition is expected to peak near 130°C. The quenched state has a major transition at 75°C and a possible shoulder at 45°C. The possible shoulder was not apparent in the DSC traces (Fig 1). These results are indicative of distinct segregation within the crystalline region of the annealed material and a possible bimodal lamellae distribution in the quenched.

Similar results have previously been attained which showed that both quenched and annealed HDPE had two transitions [13]. The quenched
transitions were at about 45 and 70°C, while those of the annealed were at 60 and 115°C. Since the higher temperature peak shifted to higher temperatures as the annealing temperature increased, the transition was attributed to relaxations in the thickened lamellae, while the low temperature peak was attributed to relaxation which occurred in the lamellae formed on cooling to room temperature [13]. Others have assigned the multiple α transitions to motion within the crystal and at the lamellae boundaries [14]. In figure 2, the quenched and annealed spectra resemble the spectra of LDPE and HDPE, respectively, as schematically illustrated by Ward [14]. This comparison is of interest because in general HDPE is less crack resistant than LDPE.

The cryogenic transitions show fewer differences between the two thermal histories. The β transition is slightly higher for the annealed case because the side chains have been worked out of the crystal lattice and are thus more free to move, however, the transition is more distinct in the quenched spectra. The amorphous transition (γ) appears broader for the annealed condition.

By measuring the area of the load-displacement hysteresis loop during the fatigue test, the dissipative capability of the materials during fracture can be obtained. This measure, W, the irreversible work, includes processes which occur in the damage zone at the crack tip, as well as in the bulk. Figure 3 shows that W is greater for the quenched state that it is for the annealed. This result agrees with the tan δ measurements, which also showed the quenched to be more dissipative. In short, the quenched material displays more plastic behavior than does the more elastic annealed material.

Ideally, one would like to view the nascent morphology, watch it develop into a damage zone, and then observe the failure of the damage zone.
Although the last step will be discussed in detail, microscopy of the first two stages was more illusive. Unfortunately, this polyethylene did not develop a spherulitic morphology, which would have been simpler to identify and quantify. Differences between the two thermal histories were imperceptible with optical microscopy. Thus, transmission electron microscopy was employed to probe differences in the lamellar morphology. A carbon replica of the etched annealed polyethylene shows rather thick (\(\sim 500\,\text{Å}\)) lamellae (Fig 4a). Below the thick lamellae region is a slightly darker area composed of much thinner (\(\sim 200\,\text{Å}\)) lamellae. This type of multiphase morphology was predicted by the DSC and DMS results. On the other hand, no similar distinguishable features could be located in the quenched material (Fig 4b).

DSC, DMS, and TEM have shown that a multiphase system develops upon annealing this ethylene/hexene copolymer. The chains reorient to form thick nonspherulitic lamellae, from which the short and highly branched chains are excluded. As will be seen in the next section, this annealed system is less resistant to fatigue crack propagation than is the more homogeneous quenched system.

**Fatigue Crack Propagation Kinetics**

Since interest lies in determining the effect of morphology, which is altered by thermal history, on fatigue crack propagation, a careful look at the fracture behavior of both thermal histories is now in order. Under identical loading conditions, the annealed material initiated and propagated a crack in a third of the number of cycles that was required for the same events in the quenched material (Fig 5). As the crack propagated, the initially brittle fracture underwent a gradual transition to a more ductile failure process. The crack length at which this transition occurred was shorter for the quenched
condition \((a/w \sim 0.5)\) than it was for the annealed \((a/w \sim 0.6)\). As a result of this transition, the specimen lifetime (number of cycles to failure \(= N_f\)) may be divided into three regions: cycles to initiate a crack \(N_i\), cycles during brittle cracking \(N_b\), and cycles during ductile cracking \(N_d\). Crack initiation is defined as 0.25 mm of crack growth from the notch, and \(N_f\) is equal to the sum of \(N_i\), \(N_b\), and \(N_d\). From figure 6 it is apparent that annealing reduced \(N_i\), \(N_b\), and \(N_f\) to about one third of the values recorded for the quenched condition. However, since the brittle to ductile transition was at a shorter crack length for the quenched state, and since the extent of ductile cracking was minimal in the annealed condition, \(N_d\) for the quenched material is more than an order of magnitude larger than that for the annealed.

The inverse slope of the curve in figure 5 is equivalent to the crack speed \((da/dN)\). Often fatigue data is displayed in a Paris plot, \(\log da/dN\) versus \(\log \Delta K\), where \(\Delta K\) is the stress intensity factor range \((K_{\text{max}} - K_{\text{min}})\). The Paris plot shown in figure 7 demonstrates a subtle difference in the kinetics of crack propagation between the two thermal histories. The quenched crack propagates at a slower rate; this rate steadily increases as the crack travels from the brittle regime (filled symbols) to the ductile regime (unfilled symbols). The annealed crack propagates at a faster rate which also steadily increases until an \(a/w\) ratio of about 0.475. In this area, the crack acceleration slows. At approximately an \(a/w\) of 0.55, the crack acceleration rate again quickens and continues to do so through the ductile regime to final failure.

In Paris plots, the data shortly after initiation to shortly before final fracture, regime II, is usually fit with a line. However, the regime II annealed data displays a definite bilinear trend. The quenched data may also display bilinear behavior, although to a much lesser extent. Regardless, a good linear
fit to the regime II data is not possible. The differences between the two thermal histories, as well as the bilinear behavior of the annealed condition, can be explained by a careful look at the mechanism of fatigue crack propagation.

**Fracture Surface**

Fatigue fracture is a discontinuous process which involves damage formation and subsequent crack advance into the damaged material [8,15]. The locations at which the crack arrests are marked by lines on the fracture surface (Fig. 8). These arrest lines, which are indicative of a discontinuous brittle fracture mechanism, are more numerous and extend across more of the specimen width in the annealed state. Whereas the arrest lines on the quenched fracture surface are composed of torn material which had been elongated at the crack tip, the arrest lines on the annealed fracture surface are macroscopically flat (Fig. 8). As the crack grows, the crack tip stress intensifies and the amount of material deformation increases. This gradual transition from brittle cracking to ductile tearing is marked by an increase in the roughness of the fracture surface and occurs at shorter crack lengths in the quenched state ($a/w \sim 0.5$) than in the annealed ($a/w \sim 0.6$). Finally, when the load bearing ligament becomes too small to bear the stress, it will yield to failure. Since brittle fracture in polymers is characterized by flat fracture surfaces and small crack tip damage zones, the fracture surfaces definitely indicate a more brittle fracture in the annealed state.

On a microscopic level, other differences between the two thermal treatments may be noted. Within a discontinuous growth band in the quenched material, the fracture surface appears relatively flat. Yet, within an annealed discontinuous growth band, there is a transition from an initially
rough area containing broken fibrils to a much smoother area near the end of the band. At larger crack lengths this intraband transition disappears, and the entire arrest band is smooth. Higher magnification of the rough texture in the short crack length arrest bands shows the surface to be covered by an open cell network which is indicative of a fracture process which involved large voids and thick well spaced fibrils (Fig 9). These type of features are typical of slow moving brittle fractures in polyethylene [15-17].

On the fracture surface of the quenched material, no such open cell pattern is formed. Instead, the entire band appears quite flat (Fig 10). Higher magnification of the band reveals lines which are one to five microns wide and oriented perpendicular to the crack propagation direction (Fig 11). Features very similar to this appear in the latter half of the short crack length arrest bands, and across the entire fracture surface at larger crack lengths, in the annealed specimen (Fig 12).

At first these observations may seem incongruous with the crack propagation kinetics. Typically, rough fracture surfaces are associated with more ductile, slower fractures. In this case, the microscopically smooth fracture surface (Fig 7) belongs to the more crack resistant specimen. This apparent anomaly will be explained after examination of the damage zone profiles. For now, it is sufficient to recall the scale of each observation. On a microscopic level there seems to be an irregularity, however, on a macroscopic level, the specimen with the roughest, most ductile, fracture is also the specimen with the slowest crack propagation rate.

Crack Tip Damage Zones and Crack Profiles

Differences between the two thermal histories can also be noted by observing the crack profiles (Fig 13). The crack tip damage zone consists of a
thick root craze, flanked by two or more side crazes. These flanking crazes are observed in small groups. Their location corresponds to that of the arrest lines on the fracture surface, and thus it is concluded that they are formed upon crack arrest [8,18].

It is generally believed that larger plastic zones result in higher resistance to crack propagation. However, as can be seen from figure 13 the amount of damage is approximately equivalent for the quenched and annealed conditions with cracks of equivalent length (a/w ~ 0.425). An approximate measure of the volume of damage material showed the annealed had 0.22 mm$^3$, while the quenched contained 0.25 mm$^3$. It seems unlikely that this minor difference in damage volume would effect the large variance in crack propagation resistance. Thus, there must be a difference in the intensity of damage within the crack tip plastic zone.

Some idea of the intensity and local deformation processes within the crack tip damage zone can be deduced by examining the crack trajectory. From figure 13, the increased tortuosity of the quenched crack path is apparent. Whereas the crack path in the annealed material is straight, that in the quenched case makes its way through tough ligaments and around other obstructions; the quenched crack tip has been deflected during propagation. The remnants of the tough ligaments can be seen in the crack profile near the notch (Fig 13), as well as on the fracture surfaces (Fig 8).

Higher magnification SEM reveals the origin of this crack tip deflection and hence toughness in the quenched condition (Fig 14). For both thermal histories small crazes occasionally develop slightly removed from the craze tip (Fig 13-14). Generally, such features "shield" the crack/craze tip from the applied stress and hence increase the material toughness. Whereas these
features are present in both the quenched and annealed specimens, they are utilized differently in each case. As can be seen in figure 14, the crack trajectory in the annealed specimen is unaffected by the presence of these small crazes. However, the growing craze tip in the quenched specimen has altered its path to propagate through the offset craze (Fig 14). As a result, the quenched damage zone has a more branched appearance and a more tortuous crack path.

This crack tip deflection can also be observed in the side crazes. Figure 15 shows that while the flanking side crazes in the annealed specimen converge, those in the quenched specimen branch. This more branched structure better distributes the applied stress and thus helps to reduce the stress intensity at the crack tip.

The origin of the tortuosuosity of the quenched crack path can be traced to the microstructure of the root craze. Figure 16 magnifies corresponding regions of the quenched and annealed root crazes which are shown in figure 14. These magnified views clearly show that the root craze in the annealed specimen is composed of large fibrils (0.5-5um) separated by gaping voids. These voids appear interconnected and thus provide a crack path of minimal resistance. On the other hand, the quenched root craze microstructure (Fig 16b) is a more continuous yielded structure. The fibrils are smaller and very closely spaced, often being laterally connected; the large voids are missing. Since the root craze in the annealed specimen already exists as a voided and relatively weak structure, the crack need not deviate from the path set by the root craze. Thus, the slightly offset side crazes have little effect because of the weakened state of the root craze. However, in the quenched condition, the would be root craze fibrils are all tied together to form a single tough unit. If
an offset craze develops nearby, the crack may alter its initial direction to propagate through this slightly weakened material (Fig 14). Such behavior is the origin of the tortuousness seen in figure 13.

DISCUSSION

Fatigue Crack Propagation Mechanism

The weakened structure of the root craze explains the origin of the anomalous fracture features discussed earlier. Due to the rounded crack tip, the advancing crack front is a free edge and thus is under plane stress. Because of this stress state, material will flow to form a continuous tough membrane along the crack front [8,19]. The flanking side crazes help to shield this crack tip region from the applied stress. In addition, for a rounded crack tip the area of highest triaxial stress is some distance ahead of the crack tip [20]. In the annealed specimen, these factors and the weakened root craze combine to cause crack advance to commence from a point within the root craze (Fig 17). The damage zone actually "unzips" in both directions from a midpoint in the weakened root craze. Because the crazed material directly ahead of the crack tip fails later, it becomes more fibrillated and voided than the material at the front of the craze tip which failed earlier. Such an unzipping phenomenon explains the transition seen within the short crack length arrest bands from a microscopically rough surface to a smoother surface. The rough area, which is composed of the open cell morphology that typifies long term failures, appears ductile on a microscopic level because it had sufficient time to void and fibrillate before it failed. The smooth area failed first and thus did not have sufficient time to fibrillate and void. Similar mechanisms and fracture surface features have previously been observed in other materials and under constant load conditions [15,19]. Such a mechanism
is also consistent with the observations of our past study in which it was assumed that the crack jump originated from the crack tip [8].

This damage zone unzipping mechanism also explains the other features of the annealed fracture surface. From figure 8a, it is apparent that the amount of microductility continually decreases during brittle crack propagation. As stated in the previous section, this microductility, caused by voiding and fibrillation, is similar to long term brittle failures of polyethylene [16-17]. In order for this type of surface feature to develop, the material must remain strained for a significant period of time. As the crack accelerates, the fracture process occurs too quickly and these features do not have time to develop. Instead, the craze fractures prior to void and fibril formation.

For the same reason, little microductility is observed in the center of the second and third arrest bands on the annealed fracture surface (Fig 8a). Because of the plane stress edge effect, the arrest bands are curved; the crack propagates faster in the center of the specimen. The increased stress concentration accelerates failure in this region as compared to the regions which are closer to the edge. This preferential center cracking has been document by microscopic observation of the rupturing crack front membrane during fatigue testing [21]. As a result of this variance in crack speed through the specimen thickness, the partially unzipped structure of figure 17 does not form at the same instance along the entire crack front. These conclusions indicate that the appearance, or lack of, microductility on the fracture surface is indicative of the local fracture speed. The fibrillated open cell network is formed by a slowly propagating crack, while faster crack speeds produce the smoother fracture surface.

From the above discussion, it may be concluded that the smooth
fracture surface of the quenched material corresponds to relatively quick fracture. On a very localized, per crack jump basis, this conclusion is true. However, because of the strength and continuity of its damage zone, the time between these crack jumps may be quite long. Unlike the annealed condition, the quenched material does not void and fibrillate because its would be root craze fibrils are all interconnected. The lack of voiding results in the root craze being able to maintain load for much longer time periods. When the damage zone does fracture, it most likely does so in a rapid continuous motion without voiding, which produces the smooth fracture surface. Thus, the time for the crack to jump from one location to the next may be quite long, but once initiated, the local fracture of the root craze is quite rapid.

The resulting microscopically smooth but finely striated surface is similar to that seen in the fast fracture areas of the annealed specimen (Figs 8-9). A very similar microstructure was observed in another MDPE, LDPE, and a nylon [16,22-23]. The origin of these striations can be postulated from the existing micrographs. The upper right corner of figure 7 shows the crack initiation site within the first band of the quenched material. From this site, which appears to be several small voids, the striations fan out. Figure 8 shows in more detail how they spread around obstructions. When there are no obstructions or nearby initiation sites, the striations are oriented perpendicular to the crack propagation direction. The position of these bands is identical to the orientation expected of a propagating crack front. Even though the crack is locally moving very fast through the discontinuous crack growth band, there is always a state of plane stress along the crack front. This stress state orients material along the moving crack front. However, since the crack is moving so quickly, the oriented material does not have time to fibrillate and void before it
is fractured. Thus, the rapidly moving local fracture leaves "tracks", which consist of a series of barely yielded membranes. Each of the membranes is curled back on itself as the crack passes it (Fig 18).

**Effect of Heterogeneous Microstructure**

It seems that we have uncovered an apparent anomaly. The annealed multiphase system is less fracture resistant than is the quenched homogeneous system. Usually, as in the case of HIPS or ABS, the addition of the second phase improves fracture resistance. Improvements in fracture toughness have even been accounted for by soft included second phases in LLDPE [24]. Second phase induced toughness is usually caused by a delocalization of damage at the crack tip in which interactions between local damage forms (craze and shear bands) diffuse the stress concentration away from the crack tip. Such filled systems have reduced moduli and improved damping. The added soft phase also acts to reduce the thickness constraint at the crack tip [25].

These processes do not occur in the annealed material. As seen in figures 11-12, damage is very localized. Damage which does develop outside of the root craze has little effect because the root craze is so weak. The annealed, multiphase system actually has a higher modulus and reduced damping (Fig 2). Due to its higher crystallinity, the annealed material is more rigid and thus increases the crack tip constraint as compared to the quenched. In addition, the second phase in the annealed system, the crystallites are too small to act as stress concentrators. Whereas the second phases in the above mentioned systems are on the order of several microns, the crystallites in the annealed polyethylene are on the nanometer level.
Effect of Tie Molecules

The reduced fatigue crack propagation resistance of the multiphase annealed specimen may be related to its lower concentration of tie molecules, which are believed to transfer loads between the crystallites. There are two possible types of tie molecules. Tie links are molecules which have crystallized in two adjacent lamellae. A molecule may also crystallize in two different areas of the same lamellae. In so doing, it forms a "loop" in the amorphous region. If this loop becomes entangled with another such loop from a different lamellae, it is defined as a tie entanglement. Since the annealed specimen has thicker lamellae than does the quenched, the probability of a molecule extending from one annealed lamellae to the next is reduced. If these thicker lamellae are not firmly linked by molecules of sufficient length, the specimen will be more prone to interlamellar fracture [4,26]. It is theorized that longer molecules form more entanglements and direct lamellae links which "tie" the crystallites together to make a more fracture resistant material [27]. This idea was supported by fatigue tests on several other pipe resins in which it was discovered that fatigue cracking resistance increased with the weight average molecular weight (Table 2). The improvement in fatigue crack propagation resistance with increasing molecular weight has also been observed many times before [4,26,28]. With this model, the number of tie molecules is proportional to the molecular weight and inversely proportional to the lamellae thickness.

Since the concentration of tie molecules can not be directly measured, indirect computational methods must be utilized. Brown and Ward estimated the tie molecule concentration in polyethylene by relating it to the brittle fracture stress [29]. They calculated the fraction of interlamellar area covered by tie molecules, $f_t$, as,
\[ f_t = C \sigma_F - \frac{\beta E_{ISO}}{E_T - E_{ISO}} \]

where \( \sigma_F \) is the brittle fracture stress, \( E_{ISO} \) is Young's modulus for van der waal's bonds (86 MPa at 90°K), \( E_T \) is Young's modulus of the tie molecules (300 GPa), \( C \) is a stress concentration factor (c. 20-50), and \( \beta \) is a proportionality constant (c. 0.1-0.2) [29].

The number of tie molecules can also be estimated by utilizing chain dimensions and probability. The basic function employed gives the probability, \( p(r) \), of finding the endpoints of a freely jointed chain separated by a certain distance, \( r \),

\[ p(r) = a r^2 \exp(-b^2 r^2)dr, \]

where \( a = (4b^3)/(\pi)^{0.5} \), \( b = (1.5/(n l^2))^{0.5} \), \( n \) is the number of chain segments which are \( l \) units long [30].

The minimum \( r \) to form entanglements is that which gives a radius of gyration \( (R_g) \) greater than half the long period \( (L_p) \), while a radius of gyration greater than the long period is needed to form links. Yeh and Runt used this idea to calculate the number of entanglements as [4],

\[ T_2(M) = \int Na' r^2 \exp(-b'^2 r^2)dr, \]

where \( T_2(M) \) equals the number of entanglements, \( N \) is proportional to the hydrodynamic volume, \( a' = (4b'^3)/(\pi)^{0.5} \), and \( b' = (1.5/(C'n l^2))^{0.5} \). C', which accounts for the limited bond angle flexibility, is 6.8; \( n \) is the number of bonds in the chain, while the bond length \( (l) \) is 1.54Å. The number of tie links was calculated in a similar fashion, however, since the number of entanglements is several orders of magnitude greater than the number of links, only entanglements were considered [4].

Yeh and Runt divided their molecular weight distribution into fractions and performed the above calculation for each fraction. In so doing, it was
shown that approximately 90% of their molecules were too short to form ties. For the fractions with nonzero \( T_2(M) \)'s, the percentage of the total distribution composed of the fraction was multiplied by the resulting \( T_2(M) \). These proportionate \( T_2(M) \)'s were then summed to give a relative measure of the number then summed to give a relative measure of the number of entanglements per molecule. Their data showed an increase in fatigue crack resistance with an increase in the number of entanglements.

Using equation 1, the number of entanglements was calculated for our quenched and annealed specimen. However, since the molecular weight distribution in the annealed specimen is exactly the same as the molecular weight distribution in the quenched specimen no effort was made to fractionate the distribution into its components. Rather, the weight average molecular weight was used to calculate \( T_2(M) \). As a result, our \( T_2(M) \)'s are an order of magnitude larger than Yeh and Runt's. In either case, the numbers may only be considered as a relative measure of tie molecules. Using this method, \( T_2(M) \) for the quenched and annealed was 152 and 96, respectively. The 1.5 times increase in the number of entanglements corresponds to a three fold increase in fatigue lifetime.

Although tie molecules may have a very significant effect on fatigue crack propagation resistance, they are not the only controlling factor. The material must have sufficient crystallinity in order to support the applied load without undergoing severe deformation. The individual crystallites must be thick enough and free of defects to maintain the load without shearing. As mentioned, the crystallites, which contribute to the needed stiffness, must be linked to prevent interlamellar failure. In order for these links to be formed, long molecules are needed. However, a high molecular weight material is
typically more difficult to process, and the deleterious effect of any processing
induced flaws may extinguish the benefits of the higher molecular weight.
Often wider molecular weight distributions are employed to avoid such
processing difficulties, however, this wider distribution also enhances the
possibility of phase segregation. Thus, each of these factors plays a role in
determining the fracture properties of the ensuing product.

CONCLUSIONS

Annealing an ethylene/hexene copolymer severely reduced its fatigue
crack propagation resistance. Although the overall fracture mechanism in both
the quenched and annealed specimens is the same, the local fracture of the
root craze is preceded by voiding and fibrillation in the annealed but not in the
quenched specimen. This root craze voiding contributes to the reduced fatigue
crack propagation resistance of the annealed specimen. The annealed
condition develops a multiphase morphology on annealing, whereas the
quenched specimen has a homogeneous microstructure. Because of its thicker
lamellae, the probability of forming tie molecules in the annealed specimen is
reduced. This reduction in tie molecule density is believed to be a major factor
in the reduced fatigue crack propagation resistance of the annealed specimen.

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### TABLE 1.1:  
**POLYETHYLENE/HEXENE X-RAY AND DSC DATA**

<table>
<thead>
<tr>
<th></th>
<th>%cry(DSC)</th>
<th>%cry(WAXD)</th>
<th>L(Å)</th>
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<tr>
<td>Quenched</td>
<td>49.0</td>
<td>49.7</td>
<td>160</td>
</tr>
<tr>
<td>Annealed</td>
<td>54.6</td>
<td>57.8</td>
<td>270</td>
</tr>
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### TABLE 1.2:  
**EFFECT OF MOLECULAR WEIGHT OF FATIGUE CRACK GROWTH**

<table>
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<th>Polyethylene</th>
<th>Comonomer</th>
<th>MW (g/mole)</th>
<th>$N_f-N_g$(cycles)</th>
</tr>
</thead>
<tbody>
<tr>
<td>F</td>
<td>butene</td>
<td>210,000</td>
<td>230,000</td>
</tr>
<tr>
<td>N</td>
<td>hexene</td>
<td>192,000</td>
<td>150,000</td>
</tr>
<tr>
<td>E</td>
<td>methylpentene</td>
<td>153,000</td>
<td>110,000</td>
</tr>
<tr>
<td>D</td>
<td>hexene</td>
<td>123,000</td>
<td>50,000</td>
</tr>
</tbody>
</table>
Figure 1.1: DSC traces of the quenched and annealed specimens.
Figure 1.2: Dynamic mechanical spectra of the quenched and annealed specimens.
Figure 1.3: Irreversible work versus crack length.
Figure 1.4: TEM of quenched and annealed morphologies (10,000x).
Figure 1.5: Number of cycles versus crack length during fatigue crack propagation test. The arrows mark the brittle to ductile transition. The crack propagation direction is from left to right.
Figure 1.6: Comparison of number of cycles to initiate ($N_i$), number of cycles of brittle cracking ($N_b$), number of cycles of ductile cracking ($N_d$), and total number of cycles to failure ($N_f$).
Figure 1.7: Crack speed versus stress intensity factor range. Filled symbols are brittle propagation; unfilled symbols are ductile propagation.
Figure 1.8a: Scanning electron micrograph of annealed fracture surface. The crack propagation direction is from left to right. (19x)
Figure 1.8b: Scanning electron micrograph of quench fracture surface. The crack propagation direction is from left to right. (19x)
Figure 1.9: Open cell microstructure seen in the arrest bands of the annealed specimen. The crack propagation direction is from top to bottom. (200x)
Figure 1.10: Microstructure of arrest band in quenched specimen. The crack propagation direction is from top to bottom. (200x)
Figure 1.11: Microstriations of the otherwise microscopically flat fracture surface of the quenched specimen. The crack propagation direction is from top to bottom. (750x)
Figure 1.12: Microstriaion seen at larger crack lengths on the fracture surface of the annealed specimen. The crack propagation direction is from top to bottom. (750x)
Figure 1.13: SEM of crack profiles and accompanying damage. The crack propagation direction is from left to right. (45x)
Figure 1.14: SEM of crack tip damage zone. The crack propagation direction is from left to right. (130x)
Figure 1.15: SEM of side crazes. The crack propagation direction is from left to right. (400x)
Figure 1.16: SEM of annealed (top) and quenched (bottom) root craze microstructure. The crack propagation direction is from left to right. (1000x)
Figure 1.17: Optical micrograph of a crack "step" in progress. The crack propagation direction is from left to right. (12x)
Figure 1.18: High magnification SEM of microstriaations seen in figure 12. The crack propagation direction is from top to bottom. (5000x)
APPENDIX 1:

PROPOSED IMPROVEMENTS IN CALCULATION OF TIE MOLECULES

In the tie molecule calculation outlined above the probability function is integrated from 1.22 L_p or 2.45L_p for entanglements or links, respectively, to infinity. However, in reality, the farthest point cannot be more than the extended chain length. This distance, rather than infinity, should be used as the integral’s upper limit (Fig A1).

If both points are within the lamellae, a tie molecule will not form. Huang and Brown excluded this possibility by multiplying their integral by one third, thereby excluding two of three dimensions occupied by the lamellae [30]. Yeh and Runt made no such correction for the number of entanglements [4]. The correct factor is dependent on L_p and r. For example, the integrated function calculates the probability of finding a point between two concentric spheres (Fig A1). The inner sphere has a radius which is proportional to L_p. The outer sphere has a radius equivalent to the length of an extended chain. The lamellae may be considered as a disk of thickness L_p and radius of an extended chain which is concentric with the spheres. To correctly account for the condition when both endpoints are in the lamellae, a factor equivalent to the percentage of the outer sphere which is not occupied by the disk must be included in the integral. This adjustment would change the function to,

\[ T_2(M) = \int N a' r \exp(-b' r^2) \left(1-\frac{L_p \pi r^2}{1.33 \pi r^3}\right) dr \]

In addition, another factor should be worked into the calculation. That factor should account for the increased probability of forming a tie molecule as the angle between the lamellae and the protruding molecule increases. If the molecule protrudes perpendicular to the lamellae, it has a greater chance of
forming a tie than if it is oriented nearly in the plane of the lamellae (Fig A2). In addition, this factor should include the increased chance of longer chains forming tie molecules. Thus far, the integral has only calculated the probability of finding points which are separated by a distance great enough to make a tie molecule. This additional factor would be proportional to \( r \) and to the sine of the angle between the lamellae and the molecule, so that the entire integral now takes the form,

\[
T_2(M) = \int \int N a' r \exp(-b' r^2) \left(1 - L \frac{\pi r^2}{2.33 \pi r^3}\right)f(r, \sin \theta) d\theta dr
\]

The last idea which should be considered when trying to quantify the effect of tie molecules on fracture resistance is the relative contribution of each type of tie. Yeh and Runt disregarded links because the number of entanglements was much greater [4]. Brown calculated links and ignored entanglements [29]. If the ties are considered as ropes, a knotted rope is typically less strong than an unknotted rope. If such an analogy is appropriate, then the entanglements contribute less per tie than do the links. In addition, the contribution of either is limited unless the ties are taught. Keith showed all intercrystalline links to be taut [31]. However, under different processing conditions loose ties may form. Relaxed links and loose entanglements would not be load bearing. As a result, they would have a reduced effect on the polymers fracture properties. Thus, it may be necessary to multiply the above function by the probability of the tie being taut. At the present time, it is unclear as how to best include this last factor.

Since direct measurements of tie molecules are not possible, attempts such as that just outlined have been made to try and estimate the probability of forming ties. Even so, there are many variables which must be considered. If
these variables can be sorted and summed in an adequate way, it may be possible to get a relatively sound estimate of the number of links and entanglements. However, the effect of such ties is still not known for certain.
Figure A1: Schematic of tie molecule formation.
Figure A2: Schematic defining theta in the proposed tie molecule concentration calculation.
CHAPTER 2:
DETERMINING FRACTURE TOUGHNESS
OF POLYETHYLENE FROM FATIGUE

ABSTRACT

A newly developed fatigue method of determining $J_{1C}$ was tested on single edge notch tension specimens of a high density polyethylene (HDPE). The fatigue method is able to propagate plane strain brittle cracks in thin specimens which more closely approximate the "in-use" thickness, and hence "in-use" morphology, of the polymer. This newly developed fatigue method was compared to ASTM standard procedure E813, which employs thick single edge notch bend specimens. The $J_{1C}$ values for both methods were within the range of HDPE $J_{1C}$ values listed in the literature. The more conservative value of the fatigue method ($J_{1C}$-ASTM = 1.8 kJ/m$^2$; $J_{1C}$-fatigue = 1.4 kJ/m$^2$) was a result of its slightly smaller damage zone.
INTRODUCTION

Load bearing polymeric components are subject to a host of stresses which may vary in intensity and frequency over the lifetime of the structure. As a result of these varied stresses, small brittle cracks may initiate from surface scratches or internal flaws and slowly propagate. Eventually, these small propagating cracks will lead to the failure of the component. These brittle cracks are characterized by very small damage zones at the crack tip and macroscopically flat fracture surfaces. While these polymers may develop brittle cracks during use, in simple monotonic tension tests, the material can display a gradual yield and high elongation at break - ductile behavior. Thus, an in-depth understanding of the resistance of ductile materials to brittle crack initiation is critical to their use in engineering structural components which experience low stresses over extended periods of time.

One method which is commonly used to test the resistance of a ductile polymer to plane strain brittle crack initiation is the ASTM standard procedure E813 - $J_{1C}$, A Measure of Fracture Toughness [1]. Several attempts to measure $J_{1C}$ of polymers have been made using E813, modifications of E813, and other J-integral methods. When alterations to the standard are limited, values of $J_{1C}$ for high density polyethylenes (HDPE) vary from 0.2 to 2.5 kJ/m$^2$ [2-5]. $J_c$ values for HDPE specimens in plane stress have been reported as high as 185 kJ/m$^2$ [6]. In one case, a $J_{1C}$ value of 99.5 kJ/m$^2$ was reported for an ultra high molecular weight polyethylene (UHMWPE), but this high value was also believed to be caused by localized plane stress conditions at the crack tip [7]. $J_{1C}$ is also sensitive to the strain rate of the experiments. Varying the strain rate has caused HDPE $J_c$ values to vary from 1.6 to 40 kJ/m$^2$ [8]. In the latter study, the ranking of different HDPE's, according to there $J_c$
values, changed when the strain rate was varied [8]. Thus, for one type of material rather large fluctuations in \( J_{1C} \) have been recorded. Similarly, very large fluctuations have been noted in \( K_{1C} \), a fracture toughness parameter which is related to \( J_{1C} \) [9].

In addition to the effects of varied strain rates, different laboratories may employ dissimilar precracking or notching methods, as well as different \( \Delta a \) measurement techniques. Variance in any of these factors can alter the \( J_{1C} \) value obtained. As a result, a great deal of caution must be exercised when comparing \( J_{1C} \) values reported from different laboratories. From a brief literature survey, it seems that many polymers have \( J_{1C} \) values in the same approximate range, i.e., 1-10 kJ/m\(^2\) [5,10-17]. The relatively small range of polymeric \( J_{1C} \) values, as well as the many factors contributing to scatter of any one value, can make material differentiation on the basis of \( J_{1C} \) difficult. However, some intralaboratory material comparisons have been able to use \( J_{1C} \) to distinguish between different polymers [4,18].

This paper reexamines the use of ASTM standard procedure E813 and introduces a new procedure, based on fatigue crack propagation, which permits the calculation of \( J_{1C} \). The ASTM method uses thick monotonically loaded specimens to determine the value of the J-integral at initiation, \( J_{1C} \). For some materials, E813 works very well. However, some tough resins do not initiate a brittle crack under the monotonic ASTM loading conditions, rather they fail by a ductile yielding process. In addition, the very thick specimens required by ASTM to produce plane strain may have significant morphological differences from the actual thinner components into which the polymer is being molded. Since fracture resistance is affected by morphology, these differences may result in differences in fracture toughness between the specimen and the actual
component.

These limitations can be overcome by our newly developed fatigue method of determining $J_{1C}$. This method is simply a modification of the ASTM E813 single specimen technique [1]. The ASTM single specimen technique employs periodic unloading of the specimen in order to determine crack lengths during crack growth from the slope of the unloading curve (compliance method). This technique is essentially a low frequency, high $R$ ratio fatigue crack propagation test. We have extended this idea by conducting a higher frequency, low $R$ ratio test on single edge notch specimens. Our modified method produces $J_{1C}$ values which are similar to those obtained using the ASTM method, but it is able to produce brittle fractures in thin specimens of very tough materials.

EXPERIMENTAL

ASTM Method

Using thick compression molded single edge notch bend (SENB) specimens, the American Society of Testing and Materials (ASTM) Standard Method E813 [1] was followed to determine the $J_{1C}$ value of a high density polyethylene (HDPE). The SENB specimens were cut from twenty seven millimeter thick compression molded plaques provided by Quantum Chemical Corporation. The specimen dimensions are shown in Figure 1.

The specimens were fatigue precracked from the seventeen millimeter deep machined v-notch until the fatigue precrack at mid-thickness was nineteen to twenty-seven millimeters long. The load during precracking cycled from 30-300 pounds at a frequency of 0.5 Hz. After fatigue precracking, the specimen was allowed to relax for one day to minimize the residual stress effects of the precracking procedure. The actual test entailed monotonically
loading the specimen at one millimeter per minute until it was judged that crack growth had occurred. At that point, the specimen was unloaded at one millimeter per minute. The load-displacement plots were recorded during the monotonic load and unload periods.

In order to observe the crack tip damage zone and to ascertain the crack tip location, the unloaded specimens were cut in half along their mid-thickness. From one half, thin sections were removed for transmission optical microscopy of the crack tip region. The other half of the specimen was fractured at a high rate to reveal the fracture surface features. Thus, the crack extension, Δa, was determined with a high degree of accuracy. The respective energy release rate was computed from the load-displacement curve (sec. 3.1 & 3.2).

Fatigue Method

Because of differences in morphology between thick specimens (ASTM) and the thinner components that the polymer is usually used in, it would be desirable to be able to determine $J_{IC}$ from specimens which approach the actual "in-service" thickness. In other words, in order to more accurately predict the fracture toughness of a loaded polymer component, a fracture toughness test should use specimens which are about the same size as the component, i.e., less than five millimeters thick. Therefore, from the same thick compression molded plaques of HDPE from which the ASTM specimens were cut, four millimeter thick single edge notch tension specimens (SENT) were prepared.

The SENT specimens were notched by pressing a razor blade to a five millimeter depth; specimen dimensions are shown in Figure 1. When properly fixed in the servohydraulic testing machine, sixty millimeters of specimen remained between the grips. The SENT specimens were fatigue loaded using
a 0.5 Hz sinusoidal waveform to minimize hysteretic heating. The zero minimum load simplified the potential energy calculation by eliminating the need for extrapolation to the x-axis (sec. 3.1 & 3.2). The $\sigma_{\text{max}}$ value used in this study was 8.2 MPa, which is about 30% of the material yield stress. Stress levels of this percentage of the yield stress have previously been shown to produce brittle failures in reasonable testing times [19]. Load-displacement hysteresis loops were recorded on an x-y plotter as the crack propagated. The crack tip position was monitored with a travelling optical microscope. Delta a was calculated as the difference between the growing crack length and the notch length.

RESULTS AND DISCUSSION

The J Integral

The $J$ integral is a path independent integral proposed by Rice [20] which describes the stress and strain fields at a crack tip. The magnitude of the $J$ integral at crack initiation, $J_{1C}$, is considered a measure of the fracture toughness of the material. $J_{1C}$ is most commonly determined using the ASTM standard test method E813 [1], which was originally designed for metals, and it is now being sought for use with polymers. This method entails monotonic loading and unloading of the specimen, and the subsequent determination of $J$ from load-displacement plots.

Because many plastics, display ductile behavior, i.e., large scale yielding, under monotonic loading, it is necessary to make very thick specimens to constrain the plasticity at the crack tip and thus produce plane strain conditions which initiate brittle cracks. The large specimen thickness required for the ASTM method severely restricts its usefulness, since molding such thick specimens may be difficult, and since most polymeric components are normally
too slender for direct testing.

For an elastic body, J is equivalent to the energy release rate, G, and is defined as the change in potential energy, U, with respect to the change in crack length, a, per unit thickness, B [21].

\[ J = \frac{-\,dU}{da \cdot B} \]  \hspace{1cm} (eq. 1).

For compact-tension or bending specimens J may be approximated using the area, A, under the load-displacement plot and the equation [22],

\[ J = \frac{2A}{Bb} \]  \hspace{1cm} (eq. 2),

where B is the specimen thickness, and b is the width of the stressed ligament. This calculation is illustrated in Figure 2. Equations 1 and 2 are also valid for the elastic-plastic situation, however, in such a case J may best be described as a measure of the stress-strain field near the crack tip [21]. Since J quantifies the deformation field at the crack tip, the value of J at the onset of crack initiation, \( J_{1C} \), may be used as a measure of fracture toughness.

**ASTM Method**

Following the ASTM E813 method, fourteen specimens were monotonically loaded and unloaded, and J was calculated using equation 2, as shown in Figure 2. Ideally, each specimen would have experienced a crack extension (\( \Delta a \)). However, measurable crack extension only occurred in twelve of the specimens.

After testing, each SENB specimen was cut in half at its mid-thickness plane. One half of the specimen was fractured at a high rate to reveal the fracture surface. While a section was cut from the other for examination of the crack tip profile. This sectioning technique was necessary because of ambiguous fracture surface features (Fig 3). Figure 4 shows the crack profile
and the accompanying half fracture surface. Higher magnification of the crack tip revealed a small craze ahead of the crack tip. This craze is responsible for creating the extra band seen on the fracture surface (Fig. 3,4,5). Without this type of careful microscopic analysis, it may be possible to mistakenly include the craze in the crack extension measurement, and as a result, $J_{1C}$ will be underestimated. Likewise, Narisawa and Takemori have reported the importance of positively identifying the fracture surface features before trying to measure crack extension [10].

After identifying the region of crack extension, $\Delta a$ was recorded as the maximum crack growth in this region as measured from optical micrographs. ASTM E813 recommends averaging nine crack growth measurements taken from the fracture surface. Because excessive crack tunnelling obscured the $\Delta a$ region near the specimen edges, the nine point technique was replaced by measurement of $\Delta a_{\text{max}}$. Obviously, an averaging technique would produce lower $\Delta a$ measurements and hence a steeper J-R curve (higher $J_{1C}$).

Having determine $J$ and $\Delta a$, the J-R curve was plotted along with the 0.15mm, 0.2 mm, and 1.5 mm blunting line offsets (Fig. 6). According to the standard, the data which lies between the 0.15 mm and 1.5 mm blunting line offsets should be fit with a power law regression curve. This limited data window is meant to select the region of crack growth in which the $J$-integral analysis is valid. Data which falls to the left of the 0.15 mm blunting line offset is excluded to prevent the inclusion of crack tip stretch phenomenon. The purpose of the 1.5 mm offset line is to insure that the amount of crack growth is within the zone of dominance of the crack tip singularity fields [23]. This limit on crack growth helps to assure that $J$ is a unique measure of the stress and strain fields at the crack tip. However, the 1.5 mm blunting line may be
overly restrictive for bending specimens, since some tests suggest that crack
growths of 0.1b are acceptable [23]. For these tests, the 0.1b limit would be
near a Δa of three millimeters. Figure 6 shows that the data which are within
the 0.1b limit do follow the same trend established by the shorter Δa data,
which fell in the smaller ASTM data window.

Using the present ASTM method (E813-87), the J versus Δa data is fit
with a power law curve. This power law regression fit was preceded in an
earlier standard (E813-81) by a linear data fit. The power law fit more closely
follows the actual data points, but it also tends to yield lower J_{1C} values than
did the linear fit. This earlier version also specified J_Q as the intercept of the
linear fit and the blunting line. Presently (E813-87), the intercept between the
power law fit and the 0.2 mm blunting line offset is termed the J_Q value (Fig
6). Our J_Q value does not qualify as a valid J_{1C} value because of excessive
crack tunnelling. Strict adherence to the standard requires that of nine Δa
measurements along the crack front, none differs by more than 7% from the
average. On some specimens, we observed a difference of over 15%. Thus, we
report a J_Q value of 4.3 kJ/m^2, rather than J_{1C}. This value was determined by
adherence to the standard, which assumes crack tip blunting and thus employs
use of the 0.2 mm blunting line offset.

This value was obtained using the 0.2 mm blunting line offset, which is
simply a line parallel to the blunting line and which has been shifted 0.2 mm on
the x-axis. The blunting line is described by the equation,

\[ J = 2\sigma_y \Delta a \] (eq. 3),

where \( \sigma_y \) is the material yield stress. The use of the 0.2mm blunting line offset
is meant to account for crack tip stretch effects which may be observed prior to
crack initiation, and be mistaken for crack growth. In some cases, loading of a
sharp notch may produce a blunted notch with some apparent crack growth. From the blunted notch, a sharp crack may subsequently initiate (Fig. 6 insert). The 0.2 mm blunting line offset is used to exclude the apparent crack growth due to blunting and thus approximate the location of crack initiation from the blunted notch. This blunting phenomenon is supported by some and opposed by others [4,10,24]. Later, we will describe how the test conditions may determine whether or not such a mechanism occurs.

**Fatigue Method**

Our recently developed fatigue method of determining $J_{1C}$ is similar to the ASTM single specimen technique. The ASTM single specimen technique follows the procedure of the multiple specimen technique described above, except that periodically during the loading of the specimen it is partially unloaded so that the crack length at that instant can be calculated from the slope of the unloading curve (compliance method). This method enables one to get several $J-\Delta a$ points from one specimen. Because of the periodic unloading and reloading, the single specimen technique is essentially a low frequency, high R ratio ($\sigma_{\text{min}}$, $\sigma_{\text{max}}$) fatigue test.

Previously, others have determined $J$ (using eq. 2) during fatigue tests and found that the results could be described by a Paris-type equation [25],

$$\frac{da}{dN} = C(\Delta J)^n$$

(eq. 4). This suggests that $J_1$ (or $\Delta J_1$) plays a definite role as a driving force of propagating fatigue cracks, which implies that the crack tip field, in fatigue, appears to be J-dominated. Thus, if $J$ can be determined during fatigue crack propagation, a $J-\Delta a$ resistance curve may be constructed. Upon appropriate extrapolation of such a curve to the initiation point, $J_{1C}$ may be obtained. Of course, a fatigue testing window defining $\sigma_{\text{max}}$, frequency, and specimen
configuration ought to be defined. This window will be framed by the conditions necessary to produce brittle crack initiation and propagation in a reasonable time period.

Expanding on this idea, fracture toughness can be determined by applying the exact energy definition of the J integral (eq. 1), rather than the approximation (eq. 2), to fatigue tests. According to this definition, J is proportional to the change in potential energy with crack growth, where potential energy is the area above the load-displacement curve (Fig. 7) [21]. We recorded the load-displacement hysteresis loops while fatiguing SENT specimens, which were prepared from the same plaques as those used to prepare the ASTM SENB specimens. The area above the loading curve of the hysteresis loop, is a measure of the potential energy of the specimen (Fig. 8). The reproducibility of the fatigue experiment is shown in figure 9, where the potential energy is plotted as a function of the crack length for three identical tests. Due to the scale of the measured quantities, the differences shown in figure 9 between the three tests are minute. Good reproducibility was also obtained at several different load levels [28]. The differential of the fit to each set of data will then be proportional to J (eq. 1) for that experiment. The change in crack length (Δa) during the fatigue test is equal to the crack length at any point minus the notch length.

The average J-R curve calculated from the three fatigue tests is shown in Fig 10 along with the ASTM J-R curve and the blunting line (eq. 3). The different slopes of the two J-R curves indicate a different resistance to continued crack propagation. Although crack propagation resistance is a geometry dependent phenomenon, one will note that the curves converge as the crack initiation region (small Δa) is approached. Figure 10 is similar to
behavior observed by Begley and Landes in which $J_{1C}$ was determined for two different specimen geometries [26].

**Determining $J_{1C}$**

Now, that the J-R curves have been generated for the ASTM method, as well as for the fatigue test, a $J_{1C}$ value must be located on each curve. $J_{1C}$ is the value of the J integral at crack initiation. Thus, the crack initiation point must be determined.

As previously discussed, the ASTM E813-87 standard recommends using the point at which the J-R curve intersects the 0.2 mm blunting line offset. This technique is meant to account for crack tip blunting effects (Fig. 6 insert). If the fatigue test is conducted at a higher stress level ($\sigma_{\text{max}} = 12.4$ MPa), such crack blunting prior to initiation will occur (Fig. 11). In such a case, use of the 0.2 mm blunting line offset would be appropriate. However, at the stress level that this fatigue test was conducted ($\sigma_{\text{max}} = 8.2$ MPa), blunting is minimal (Fig. 12) and therefore the use of the blunting line offset overestimates the amount of crack tip stretch prior to initiation.

These initiation processes were visible because the fatigue specimens were only four millimeters thick. For the ASTM SENB specimen, the crack initiation process could not be observed directly because the crack initiated in the center of the twenty-seven millimeter thick opaque specimen. As an alternative to observing the crack at initiation, microtomed sections were removed from the thick ASTM SENB after a crack had initiated. By comparing the crack profile of the SENB specimen to that of a fatigued SENT specimen, which had a comparable crack extension, we are able to judge the relative difference in brittleness (plane strain contribution) between the two specimens (Fig. 13). Although the SENB does have a slightly larger craze than
the fatigue SENT specimen, both cracks have a similar sharp appearance. Since it is known that blunting was minimal in the fatigue SENT specimen, we assume that it was also minimal in the ASTM SENB specimen because of the similarity in their crack tip profiles. This conclusion negates the use of the 0.2 mm blunting line offset and indicates that the initiation point for both specimens must lie to the left (smaller \( \Delta a \)) of the 0.2 mm blunting line offset.

Having disqualified the use of the 0.2 mm blunting line offset, a more accurate prediction of the crack initiation point must be made. If one can ascertain that absolutely no blunting occurred, the crack initiation point would correspond to the y-axis (\( \Delta a = 0 \)). However, since some blunting has occurred (but much less than estimated by the 0.2 mm blunting line offset) an initiation point between the y-intercept (\( \Delta a = 0 \)) and the 0.2 mm blunting line offset would be appropriate. In addition, because a power law regression fit was used for the ASTM data, choosing the y-intercept as the initiation criteria would always yield a \( J_{1C} \) of zero for the ASTM method. Thus, we believe the blunting line may be the best estimate of crack initiation for these experiments. However, under different loading conditions or different specimen geometries, it would be necessary to observe the crack initiation phenomenon for each case and then determine the appropriate location on the J-R curve which corresponds to \( J_{1C} \).

The intersection of the J-R curves with the blunting line indicates a \( J_{1C} \) of 1.7 kJ/m\(^2\) for the ASTM SENB specimen and a more conservative \( J_{1C} \) of 0.8 kJ/m\(^2\) for the SENT fatigue specimen (Fig 10). Both these \( J_{1C} \) values are within the range of \( J_{1C} \)'s (0.2 - 2.5 kJ/m\(^2\)) reported in the literature for HDPE's [2-5]. Considering that these two methods differ in specimen geometry and specimen thickness, and that the two methods use very different
loading conditions, the agreement between the two methods is excellent. However, even the small difference between $J_{1C}$ values can be explained. In comparison to our fatigue method, the larger craze of the ASTM method is indicative of greater strains at the crack tip (Fig. 13). Because $J$ is a measure of the strain field around a crack tip, this larger craze (higher crack tip strain) results in a slightly higher $J_{1C}$ value. Knowing the origin of this relatively small difference in $J_{1C}$, it seems clear that fatigue can be used to obtain $J_{1C}$ values which are nearly the same as the $J_{1C}$ values obtained from the ASTM procedure.

**Comparison of ASTM and Fatigue Methods**

In addition to producing similar $J_{1C}$ values to the ASTM method, the main advantage of the fatigue test is that it can be done with specimen thicknesses which more closely approach the thickness in which the polymer will be used. The ASTM method requires thick blocks to constrain the ductility at the crack tip and thus produce a plane strain brittle fracture, the "worst case scenario." On the other hand, fatigue can be used on much thinner specimens to produce fractures which are even more brittle (higher plane strain contribution) than those observed in the ASTM tests (Fig 13). The primary importance of this advantage is quite simple. Because of differences in thermal history and processing methods (i.e., compression molding vs. extruding), differences in morphology arise between thick test specimens and the relatively thin actual components. These morphological differences are expected to affect fracture resistance. Thus, because of differences in morphology, the fracture resistance of a thick block specimen may not be an accurate measure of the fracture resistance of the actual thin component, even though both are made of the same material. However, if one employs fatigue,
specimens may be tested which approach the same thickness, and thus morphology, as the real component. Testing these thinner specimens will give a more accurate assessment of the fracture toughness of the polymer part.

Another difference between the two methods is tied to the fact that for some tough polymers, brittle cracks can not be initiated using monotonic loading. For example, Crist and Carr have conducted J integral studies on thick compact tension specimens of tough medium density polyethylene pipe resins [27]. The "crack" which initiates is shown in figure 14. Under the conditions specified by ASTM E813, these materials separate by a tearing process which is accompanied by a large damage zone; they do not crack under monotonic load. Since we are testing for a materials resistance to brittle failure, we must have a test method which can produce a brittle crack. The SEN fatigue test can produce brittle failures in thin specimens of these tough resins, as well as provide $J_{1C}$ values [28].

From the above discussion it should be clear that fracture toughness testing of polymers is no simple task. In order to produce a plane strain brittle fracture the crack tip plasticity must be constrained. This constraint can be accomplished by monotonically loading very thick specimens or by propagating a crack under the lower loads used in fatigue. Since $J_{1C}$ is a measure of the strain field near the crack tip, varying the fatigue conditions, which changes the size of the damage zone [29], will change $J_{1C}$. For example, higher load levels will produce more crack tip damage and hence larger $J_{1C}$'s. In this case, we have selected fatigue conditions ($\sigma_{\text{max}} \sim 30\%\sigma_y$, 0.5 Hz, R = 0) which produce a damage zone which is roughly the same size as the damage zone in the ASTM test (Fig 13); as a result, both $J_{1C}$ values are similar. To our knowledge, this work is the first attempt to determine $J_{1C}$ from fatigue tests.
The present study appears promising, but further work is needed to establish an appropriate window of fatigue conditions for which the method is applicable.

CONCLUSIONS

By applying the energy definition of the J-integral to fatigue tests, a method has been developed which permits the calculation of $J_{1C}$ from thin SENT specimens. This method is capable of producing plane strain brittle failures, characterized by small crack tip damage zones and macroscopically flat fracture surfaces, in thin specimens of tough polymers. The application of this newly developed fatigue method to HDPE has yielded a $J_{1C}$ value similar to that obtained using the ASTM method ($J_{1C-ASTM} = 1.8 \text{ kJ/m}^2$; $J_{1C}$-fatigue = 1.4 kJ/m$^2$). The more conservative fatigue value is a result of less intense crack tip deformation.

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Specimen Geometries

ASTM SENB

Fatigue SENT

Figure 2.1: Geometries of the ASTM single edge notch bend (SENB) and single edge notch tension (SENT) specimens.
Figure 2.2: An actual ASTM SENB loading and unloading curve. The hatched area (A) is used to approximate the J integral. Crack growth (Δa) for this specimen was 2.9 mm.
Figure 2.3: Area on the ASTM SENB fracture surface area where crack growth occurred, showing ambiguous fracture surface features between the precrack and rapid fracture regions.
Figure 2.4: Comparison of partial ASTM SENB fracture surface and the corresponding crack profile used to identify the crack tip position.
Figure 2.5: Partial ASTM SENB fracture surface with each area clearly identified.
Figure 2.6: The J-R curve for the ASTM method. The solid line is a power law fit to the filled points. The unfilled points fall outside the ASTM data window. The straight dashed lines are the 0.15, 0.2, and 1.5 mm blunting line offsets. The insert shows the blunting phenomenon which is approximated by the 0.2 mm blunting line offset.
\[ J = -\frac{dU}{da \cdot t} \]


Figure 2.7: The area above the loading curve is proportional to the negative potential energy (-U). As the crack grows from \( a \) to \( a + \Delta a \), the area between two loading curves is proportional to the change in potential energy (\( \Delta U \)) associated with that increment of crack growth (\( \Delta a \)) [21].
Figure 2.8: Determination of $J$ from recorded load-displacement hysteresis loops.
Figure 2.9: Evolution of the potential energy as a function of crack length during three fatigue tests.
Figure 2.10: J-R curves for the ASTM and fatigue conditions.
\[ \sigma_{\text{max}} = 12.4 \text{ MPa} \]

Figure 2.11: The crack tip profile at initiation during a high load level fatigue test which shows the blunting phenomenon described in figure 6 insert.
Figure 2.12: The crack tip profile at initiation during the actual low load fatigue tests. The crack tips have been outlined.
Fatigue ($\sigma_{\text{max}} = 8.2\text{MPa}$)

ASTM E813

0.25 mm

Figure 2.13: Crack tip profiles of interrupted fatigue and ASTM tests shortly after initiation.
Figure 2.14: Crack tip profile of an ASTM test in a tough medium density polyethylene. (50x)

ref: B. Crist, et. al., GRI Annual Meeting, 9/5/91
CHAPTER 3:
CORRELATING $J_{1C}$ AND
CRACK TIP DAMAGE IN POLYETHYLENE

Abstract

The energy definition of the J-integral was used to determine $J_{1C}$ of
high density polyethylene under four different fatigue loading conditions. The
value of $J_{1C}$ increased as the maximum stress during the fatigue test increased.
A linear relationship was observed between the length of the crack tip damage
zone and the $J_{1C}$ value for each test condition.
INTRODUCTION

Nearly since its introduction [1], the J-integral has been employed as a measure of fracture toughness in materials which display small scale yielding. The value of the J-integral during plane strain crack initiation is termed $J_{1C}$, which is often viewed as a material property indicative of fracture resistance. By definition, the J-integral is a path independent line integral which provides a measure of the deformation field at the crack tip [2]. Since the deformation field (crack tip damage zone) is affected by the material, as well as the loading conditions, both factors should be considered when trying to quantify fracture toughness. Others have shown that $J_{1C}$ varies widely when different loading rates are used to test polyethylenes [3,4]. These rate induced variances are significant enough to alter the $J_{1C}$ based fracture resistance ranking of the different polyethylenes [3]. These $J_{1C}$ rate effects are a result of differences in damage development at the crack tip. Crack tip deformation in viscoelastic materials, such as polyethylene, is dependent on the rate and maximum stress during loading. Thus, this paper examines how varied loading conditions create different quantities of damage at the crack tip, which may subsequently be reflected by the $J_{1C}$ values obtained.

EXPERIMENTAL

The 100 x 27 x 4 mm high density polyethylene (HDPE) single edge notch (SEN) specimens used for this series of tests were milled from a compression molded plaque. The specimens were notched at mid-length by pressing a razor blade to a five millimeter depth.

Upon fixing the specimens in the servohydraulic MTS machine, the initial grip-to-grip distance was 60 mm. Load control fatigue experiments were conducted using a 0.5 Hz sinusoidal waveform which varied from zero stress to
the preset maximum (6.2, 8.2, 10.3, or 12.4 MPa). These loads are 23, 30, 38
and 46% of the 27 MPa yield stress (σ_y). Loads of these percentages of σ_y
have previously been shown to promote brittle failure during fatigue tests of
polyethylene [5]. Two to three experiments were conducted at each load level.

At least one experiment of each set was interrupted at a crack length of
eight millimeters so that the damage zone at the crack tip could be examined.
Ten micron thick sections were cryomicrotomed from midthickness of these
interrupted specimens. The sections were opened to the approximate crack
opening displacement which was present during the experiment, and
transmitted light optical micrographs were taken of the crack tip and preceding
damage zone.

During the experiments load versus displacement hysteresis loops were
recorded on an analog x-y recorder. The crack tip position was monitored with
a travelling optical microscope.

RESULTS AND DISCUSSION

Fracture Features

In the past, J_{1C} tests on viscoelastic polymers had to be conducted using
very thick specimens or low temperatures in order to constrain the ductility at
the crack tip and produce a plane strain brittle fracture. Plane strain fractures
are characterized by small damage zones and flat fracture surfaces. Despite
using SEN specimens which are only four millimeters thick, figures 1 and 2
show that flat fracture surfaces and small damage zones have been obtained by
employing fatigue at ambient temperature. The similarity in fracture features
between these thin SEN fatigue specimens and the thick monotonically loaded
ASTM bend specimens has previously been demonstrated [6].

As the crack propagates during our load control test, the stress on the
remaining ligament increases. As a result, the fracture process passes through a gradual brittle to ductile transition, which can be observed by the increased roughness of the fracture surface at longer crack lengths (Fig. 1). Obviously, at higher loads this transition occurs at shorter crack lengths. However, under each condition, there exists a definite plane strain brittle crack propagation region, manifest by the macroscopically flat fracture surface.

\( J_{IC} \) from Fatigue

A method for measuring \( J_{IC} \) from fatigue experiments has been developed and compared to ASTM E813 [6,7]. This fatigue method of determining \( J_{IC} \) employs the energy definition of the J-integral [8],

\[
J = -\frac{dPE}{da} B,
\]

where PE is the potential energy, \( a \) is the crack length, and \( B \) is the specimen thickness. The potential energy is proportional to the area above the loading curve in a load-displacement plot (Fig. 3). Therefore, by monitoring the change in area above the load-displacement hysteresis loops with change in crack length, the J integral can be determined for a propagating fatigue crack. Since this potential energy is typically a polynomial function of crack length, a complete resistance curve, \( J \) versus \( \Delta a \), can be obtained using this method. Delta \( a \) is the difference between the crack length and the notch depth. A similar type of \( J-\Delta a \) plot is obtained from the ASTM technique.

In either case, \( J_{IC} \), the value of the J-integral at initiation, must be determined. How this value is selected depends on the crack initiation phenomenon. If crack tip blunting occurs prior to crack initiation, use of the 0.2 mm blunting line offset is justified [6]. However, in the case of crack initiation without blunting, \( J_{IC} \) values should be selected at smaller \( \Delta a \) values.
In such cases, the intercept between the blunting line and the J-R curve is more correct [6].

**J_{1C} Analysis**

Figure 4 shows the potential energy as measured from the area above the hysteresis loops for each set of experiments. As the crack grows, the specimen becomes more compliant, and as a result, the area above the hysteresis loops increases. This gradual increase in potential energy with crack length is more noticeable at higher loads. Figure 4 also demonstrates the excellent reproducibility of the fatigue experiments. Each data set was fit with a fourth order polynomial. The derivatives of these fits are then proportional to J (eq 1). The average J-R behavior for each condition is shown in figure 5 along with the blunting line and 0.2 mm blunting line offset.

As discussed previously, $J_{1C}$ is the value of the J-integral at initiation. For cases which involve crack tip blunting, the 0.2 mm blunting line offset can be used to estimate crack initiation. For the two higher load cases (10.3, 12.4 MPa) crack tip blunting (Fig. 6) was observed prior to crack initiation. Therefore, $J_{1C}$ is estimated as the intercept between the 0.2mm blunting line offset and the J-R curves: $J_{1C}$ - 10.3 = 2.7 kJ/m$^2$, $J_{1C}$ - 12.4 = 4.5 kJ/m$^2$. For the low load cases little crack tip blunting was observed (Fig 7) prior to initiation. As a result, use of the blunting line offset would not be appropriate. Instead, $J_{1C}$ is selected as the intercept between the blunting line and the J-R curve: $J_{1C}$ - 8.2 = 1.4 kJ/m$^2$, $J_{1C}$ - 6.2 = 0.8 kJ/m$^2$.

**$J_{1C}$ & Damage Zones**

Since $J_{1C}$ varies with the loading conditions, it is obviously not a material parameter. Even using the more conventional ASTM E813 method to calculate $J_{1C}$, it has been shown that $J_{1C}$ fluctuates when the loading
conditions are varied [3,4]. However, the J-integral was never meant to be a material parameter. It is a measure of the deformation fields at the crack tip. Since the amount of deformation varies with the loading conditions, one would expect $J_{1C}$ to vary also.

Figure 2 shows the damage zones which preceded the crack at each of the four loading conditions. Each section is a ten micron thick microtomed section which was cut from mid thickness of the specimen. Each specimen contained an interrupted fatigue crack approximately eight millimeters long ($\Delta a = 3$ mm). Obviously, as the load increased so did the amount of damage. Yet, the same approximate damage zone shape is maintained.

Figure 8 compares the length of the damage zones as seen in figure 2 to the $J_{1C}$ values as estimated from figure 5. Also included in figure 6 are the results of other studies [4,9]. These authors used monotonically loaded C-T specimens and different types of HDPE. Despite the differences in geometry, procedure, and material, there seems to be an almost linear correlation between the damage zone length and the $J_{1C}$ value. In retrospect, the correlation of $J_{1C}$ to damage zone size may seem obvious, because the J integral is a measure of the crack tip deformation field [2]. However, we believe this is the first attempt to quantitatively relate the two parameters. Previously, others have shown that materials with larger damage zones have larger $J_{1C}$ values and are more resistant to fracture [9], but the size of the damage zones were never quantified. For each case, $J_{1C}$ was implied to be a material parameter. In reality, $J_{1C}$ is a measure of the size of the crack tip damage zone (Fig 6), which is dependent on the material and the loading conditions.
CONCLUSIONS

A linear relationship has been found between the length of the damage zone and $J_{1C}$ in HDPE tested under four different loading conditions. This relationship was confirmed by comparing results of authors who used different geometries, procedures, and types of HDPE. Since $J_{1C}$ depends on damage zone length, which varies with test conditions, $J_{1C}$ is not solely dependent on material. $J_{1C}$ is a measure of the crack tip damage zone size, which varies with material and testing conditions.

ACKNOWLEDGEMENTS

The financial support of the Gas Research Institute is gratefully acknowledged. The assistance of Joydeep Ghosh during data compilation is appreciated.
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Figure 3.1: Fracture surfaces of failed SEN specimens tested under different maximum loads. The crack propagation direction is from left to right.
Figure 3.2: Crack tip profiles of interrupted SEN specimens tested under different maximum loads. The crack propagation direction is from left to right.
Figure 3.3: Schematic illustration of load-displacement plots showing measurement of the negative potential energy.
Figure 3.4: Potential energy versus crack length plot for the different maximum load conditions.
Figure 3.5: J-R curves for the four different loading conditions, as well as the dashed blunting line and the dotted 0.2 mm blunting line offset.
Figure 3.6: Crack tip profile before and after crack initiation at $\sigma_{\text{max}} = 12.4$ MPa.
Figure 3.7: Crack tip profile before and after crack initiation at $\sigma_{\text{max}} = 6.2$ MPa.
Figure 3.8: Length of damage zone plotted as a function of $J_{1C}$ for the four load conditions, as well as some literature values.
CHAPTER 4:
A TIME DEPENDENT FRACTURE TOUGHNESS
MEASURE FOR POLYETHYLENE

ABSTRACT

Medium density polyethylene (MDPE) resins exhibit extraordinarily long time-to failure, due to their superior brittle cracking resistance. Attempts to quantify this phenomenon using the J integral approach have been hampered by the unrealistically large specimen thickness required to effect plane strain conditions and because monotonic loading does not readily initiate brittle cracks in MDPE resins. A procedure to determine fracture toughness (J_{1C}) using fatigue crack propagation has been developed. In addition to simplifying fracture toughness measurements, the new method produces useful information about the time dependence of crack initiation and the rate of crack propagation. The fracture toughness of a number of medium density polyethylenes was successfully determined. However, J_{1C}, by itself, is not precise enough to quantify differences in brittle crack propagation resistance between MDPE resins. By incorporating both time and energy, the factors responsible for brittle fractures of MDPEs, a measure of the power to fracture has been obtained which can differentiate between resins on the basis of their brittle cracking resistance.
INTRODUCTION

Load bearing polymeric components are subject to a host of stresses which may vary in intensity and frequency over the lifetime of the structure. These varied stresses are applied for long periods of time and may initiate small brittle cracks from surface scratches or internal flaws. Eventually, these small cracks will propagate through the structure and produce a failure. These brittle cracks are characterized by very small damage zones at the crack tip and macroscopically flat fracture surfaces.

A material which displays small scale yielding can be tested for resistance to such brittle (plane strain) crack propagation by using the American Society of Testing and Materials (ASTM) standard procedure E813 - J1C, A Measure of Fracture Toughness [1]. This method uses thick monotonically loaded specimens to determine the value of the J-integral at initiation, J1C. However, it is possible that differences in thickness between the specimen and the actual structure may induce morphology variances, which may cause the fracture resistance of the test specimen to diverge from that of the component. In addition, some tough resins do not initiate a brittle crack under the conditions of ASTM E813 [1].

This paper discusses a method which overcomes both of these difficulties. Using fatigue, we have designed a test which permits the determination of J1C from brittle crack propagation in thin specimens of tough medium density polyethylene (MDPE). Yet, because J1C neglects time, it was unable to detect differences in cracking resistance between different polyethylene resins. Thus, we propose a fracture toughness method which incorporates both energy and time, the factors inherent to brittle cracking of polyethylene.
EXPERIMENTAL

Using thick compression molded single edge notch bend (SENB) specimens, the American Society of Testing and Materials (ASTM) Standard Method E813 [1] was followed to determine the $J_{1C}$ value of a high density polyethylene (HDPE). The 204 x 51 x 27 mm SENB specimens were v-notched to a 17 mm depth and then fatigue precracked to a final depth of 19 - 29 mm.

Using the same thick compression molded plaques of HDPE from which the ASTM specimens were cut, four millimeter thick single edge notch tension specimens (SENT) were cut. The 100 x 27 x 4 mm SENT specimens were notched by pressing a razor blade to a five millimeter ( +/- 0.75 mm) depth at mid-height. The SENT specimens were fatigue loaded using a 0.5 Hz sinusoidal waveform to reduce hysteric heating. The zero minimum load simplified the potential energy calculation by eliminating the need for extrapolation to the x-axis. The $\sigma_{\text{max}}$ value used in this study was 8.2 MPa, which is about 30% of the material yield stress. Stress levels near this percentage of the yield stress have previously been shown to produce brittle failures in reasonable testing times [2]. Load-displacement hysteresis loops were recorded on an x-y plotter, and crack length measurements were made using a travelling optical microscope.

In addition to the HDPE SENT specimens which were cut from the thick ASTM plaques, four millimeter thick plaques were compression molded from four medium density polyethylene (MDPE) pipe resins and a different HDPE. SENT specimens of 145 x 20 x 4 mm, with a five millimeter deep pressed razor notch, were cut from the plaques and tested at 1 Hz with a $\sigma_{\text{max}}$ of 6.7 MPa. The fatigue method was then applied to these samples to see if
differences in fracture resistance could be quantified using $J_{1C}$.

RESULTS AND DISCUSSION

ASTM Method

Following ASTM E813, twelve identical HDPE bending specimens were monotonically loaded and unloaded, and $J$ was calculated from the approximation [3],

$$J = \frac{2A}{Bb} \quad \text{(eq. 1)},$$

where $A$ is the area under the load-displacement curve, $B$ is the specimen thickness, and $b$ is the uncracked ligament width. Maximum crack extension ($\Delta a$) during loading was measured from the bands on the fracture surface. The seven data points which fell within the ASTM data window [1] were fit with a power law curve (Fig 1). Adherence to the standard requires that of nine measurements along the crack front, none differs by more than 7% from the average. On some specimens, a difference of over 15% was observed. Since such results are indicative of excessive crack tunnelling, we report a $J_Q$ value of 4.3 kJ/m$^2$, rather than $J_{1C}$.

This value was obtained from the intersection of the power law fit and the 0.2 mm blunting line offset. The blunting line is described by the equation,

$$J = 2\sigma_Y \Delta a, \quad \text{(eq. 3)}$$

where $\sigma_Y$ is the material yield stress. The use of the 0.2mm blunting line offset is meant to account for crack tip stretch effects which may be observed prior to crack initiation, and be mistaken for crack growth. In some cases, loading of a sharp notch may produce a blunted notch with some apparent crack growth. From the blunted notch a sharp crack may subsequently initiate (Fig 1 insert). The 0.2 mm blunting line offset is an arbitrary method to exclude the apparent
crack growth due to blunting and thus approximate the location of crack initiation from the blunted notch. This blunting phenomenon is supported by some and opposed by others [4,5,6]. Later, we will describe how test conditions may affect whether or not such a blunting mechanism occurs.

Fatigue Method

Fracture toughness can also be determined by applying the exact energy definition of the J integral,

\[ J = \frac{-dPE}{d\Delta a} \]  \hspace{1cm} (eq. 2).

This definition states that J is proportional to the change in potential energy (PE) with crack growth, where the potential energy is the area above the load-displacement curve [7].

Transposing this idea to fatigue, the area above the loading curve of the load-displacement hysteresis loop is a measure of the potential energy of the specimen (Fig 2 insert). Figure 2 shows good reproducibility of the potential energy evolution. As our testing and measurement techniques are refined, the 15% variance should be reduced. The differential of the fit to the potential energy versus \( \Delta a \) data is proportional to J (eq. 2). The change in crack length \( \Delta a \) is equal to the crack length at any point minus the notch length. The J-R curve calculated in such a manner is shown in Fig 3 along with the ASTM J-R curve. The difference in the slopes and inflections of the two curves is a geometry effect which has been observed in the past [8]. Essentially, this difference indicates that crack tip damage in a tension specimen evolves differently than in a bending specimen.

\textbf{J_{1C} Criterion}

Now, that the J-R curves have been generated for the ASTM method,
as well as for our fatigue method, a $J_{IC}$ value must be identified on each curve. 
$J_{IC}$ is the value of the $J$ integral at crack initiation. Thus, the crack initiation 
point must be determined.

As previously discussed, the ASTM E813 standard recommends using the 
point at which the J-R curve intersects the 0.2 mm blunting line offset. This semi-arbitrary technique is meant to account for crack tip blunting effects (Fig 1 insert). All cracks have some crack tip curvature, and a brittle failure in a polymer would not appear brittle to a metallurgist. Thus, defining crack 
blunting and crack initiation is somewhat subjective, especially when it is recalled that this analysis is an application of a metals standard to polymeric fracture. When the fatigue test is conducted at a high stress level ($\sigma_{\text{max}} = 12.4$ MPa), crack blunting prior to initiation will occur (Fig. 4). In such a case, use of the 0.2 mm blunting line offset would be appropriate. However, at the stress level that this fatigue test was conducted ($\sigma_{\text{max}} = 8.2$ MPa), blunting is minimal and therefore the use of the blunting line offset overestimates the amount of crack tip stretch prior to initiation (Fig. 5). These initiation processes were visible from the specimen edge because the fatigue specimens were only four millimeters thick.

For the ASTM SENB specimen, which was twenty-seven millimeters thick, crack initiation processes could not be observed because the crack initiated in the center of the thick opaque specimen. As an alternative to observing the crack at initiation, microtomed sections were removed from the thick ASTM SENB after a crack had initiated. By comparing the crack profile of the SENB specimen to that of a fatigued SENT specimen which had a comparable crack extension, we are able to judge the relative differences in brittleness between the two specimens (Fig 6). Although the SENB does have
a slightly larger craze than the fatigue SENT specimen, both cracks have a similar sharp brittle appearance. Since it is known that blunting was minimal in the fatigue SENT specimen, we assume that it was also minimal in the ASTM SENB specimen, because of the similarity in their crack tip profiles. This conclusion recants the routine use of the 0.2 mm blunting line offset and indicates that the crack initiation point for both specimens must lie to the left (smaller $\Delta a$) of the 0.2 mm blunting line offset. For this series of experiments, the blunting line gives a more reasonable estimate of the crack initiation phenomenon than does the 0.2 mm blunting line offset.

The intersection of the J-R curves with the blunting line indicates a $J_{1C}$ value of 1.8 kJ/m$^2$ for the ASTM SENB specimen and a $J_{1C}$ value of 1.4 kJ/m$^2$ for the SENT fatigue specimen. Since the ASTM conditions produced a slightly larger craze (greater crack tip strain) than did the fatigue, and since $J_1$ is a measure of the deformation field around a crack tip, one would expect the ASTM method to yield a larger $J_{1C}$ value. Knowing the origin of this relatively small difference in $J_{1C}$, it seems clear that fatigue can be used to obtain $J_{1C}$ values which are equivalent to the $J_{1C}$ values obtained from the ASTM procedure. Comparison of the two methods shows that $J$ at initiation, $J_{1C}$, is independent of the test method. However, as seen by the slopes of the J-R curves (Fig. 3), $J$ during crack propagation depends on the geometry and test conditions.

In addition to producing similar $J_{1C}$ values to the ASTM method, the main advantage of the fatigue test is that it can be done with thicknesses, and hence morphologies, which are more realistic for most polymer applications. The monotonic bending test requires much thicker specimens.
\textbf{J}_{1C} \text{ of MDPE}

Even with very thick specimens, for some tough polymers, brittle cracks can not be initiated using monotonic loading. For example, Crist and Carr have conducted J integral studies on thick compact-tension specimens of tough medium density polyethylene resins [10]. Under the conditions specified by ASTM E813, these materials separate by a tearing process which is accompanied by a large damage zone; they do not \textit{crack} under monotonic load. Since we are testing for resistance to plane strain brittle failure, a test method should be used which produces brittle fractures. The SEN fatigue test can produce \textit{brittle} failures in these tough materials, as well as provide J\textsubscript{1C} values.

Fatigue lifetimes and past experience have shown MDPE’s to be much more resistant to fracture than HDPE’s. Yet, table 1 shows that a higher J\textsubscript{1C} value was obtained for the HDPE than for the MDPE’s. In addition, independent tests have shown differences in crack propagation resistance within these MDPE’s. These differences are not manifest by the J\textsubscript{1C} values, which, due to experimental uncertainty ( +/- 0.5 kJ/m\textsuperscript{2}), are all approximately equivalent. A brief literature survey of polymeric ASTM J\textsubscript{1C} testing confirms the difficulty of using J\textsubscript{1C} to differentiate materials. This sampling shows most polymeric J\textsubscript{1C} values are in the 1 - 10 kJ/m\textsuperscript{2} range [4,5,11-20]. In some cases, lab to lab variability of these J\textsubscript{1C} values can be quite wide [13,18]. Thus, using J\textsubscript{1C}, by itself, to separate polymers of varying fracture toughness may be difficult.

\textbf{Time and Energy}

As stated earlier, if polymeric structures develop brittle cracks, they are usually a result of the structure being exposed to a variety of low stresses applied over a long time period. During this time, damage zone growth
processes occur even prior to crack initiation and during crack arrest. This
time dependent damage growth consumes energy and affects the overall
fracture process (Fig. 7). However, $J_{1C}$ only quantifies the change in
potential energy during crack growth. Without crack growth there cannot be a
$J_{1C}$, yet there can be time-dependent damage accumulation and energy
dissipation (Fig 7a-b). Thus, a more sensible, and hopefully more sensitive,
measure of fracture toughness should incorporate both factors, energy and
time.

The need to incorporate time in fracture toughness tests of viscoelastic
materials has been displayed before. During a crack layer analysis of two
different MDPE pipes, it was demonstrated that both pipes developed nearly
equivalent amounts of damage [2]. From the crack layer theory, $J$ is equivalent
to the size of this damage ($R$) multiplied by a material constant, the specific
enthalpy of damage ($\gamma^*$). Despite the fact that the lifetimes of the tow pipes
differed by a factor of eight, their specific enthalpies of damage only differed
by twenty percent. Obviously then, $J$ did not give a good indication of the
difference in material toughness. This shortcoming was caused by the
differences in the damage accumulation rate. Under identical conditions,
damage evolution, and hence the subsequent crack growth, was much slower in
the tough specimen than it was in the more fracture prone specimen. This
observation underscores the need to incorporate rate effects in polymeric
fracture tests.

**Power to Fracture**

Figure 8 shows the time effect in several fatigue tests. The number of
cycles is plotted as a function of crack length during testing of one MDPE
under four different fatigue conditions. The conditions vary in their maximum
stress, frequency, and notch depth. Obviously, these major differences create very different crack propagation kinetics. As can be seen in figure 9, the potential energy during each fatigue condition evolves differently also. Since crack growth in these specimens is a discontinuous process, it is conceivable to break the crack growth into increments. By doing such, it is possible to determine the number of cycles elapsed (\(\Delta N\)) as the crack grows from \(a_1\) to \(a_2\) (Fig 8). In a similar manner, the average potential energy (\(PE_{\text{avg}}\)) of the specimen during that crack growth period can be determined (Fig 9). Multiplying \(PE_{\text{avg}}\) by \(\Delta N\) and by the frequency of the test yields the power to fracture (\(J/m^2s\)). Despite the varied fatigue conditions, figure 10 shows that the evolution of the power to fracture is relatively constant within the chosen fatigue range. All four conditions reduce to a single narrow "power band."

This "power band" was only calculated in the brittle crack propagation region. As the crack propagates, the load bearing ligament becomes smaller, the stress intensity factor at the crack tip is increased, and as a result, the ductile contribution to fracture, which is proportional to the roughness of the fracture surface, is increased (Fig. 11). This gradual brittle to ductile transition occurs near a crack length of ten millimeters. This work is an attempt to quantify brittle fracture. Since after such a transition the failure process is one of ductile tearing rather than brittle fracture, the region beyond a ten millimeter crack length has not been included in the power analysis.

Since the power to fracture is independent of our chosen test condition, it may be indicative of the intrinsic fracture resistance of the material. In order to check if the power to fracture is indicative of a material's resistance to fracture, the power analysis has been applied to four different MDPE's and a HDPE tested under the same condition (\(\sigma_{\text{max}} = 6.7\) MPa, \(\nu = 1\) Hz). Figure
12 shows that material differentiation is possible with the power analysis. The resins which are most resistant to brittle cracking display the highest power to fracture (Table 1). Figure 12 also shows that, despite the variance in fatigue conditions for the testing of the one MDPE, the resulting power band is still narrow enough to permit material differentiation.

Of course, this method does have limitations. In each experiment discussed above the crack propagation mechanism was the same. Brittle discontinuous crack growth occurred up to a crack length of about ten millimeters, after which the failure became ductile. In one case, the test was conducted at a very high stress (8.4 MPa). This condition produced a noticeably more rounded power curve. Likewise, if one were to test at very low stress levels the test would be prohibitively time consuming and the curve may deviate from the established trend. Obviously, the appropriate loading conditions are dependent on the material, since a similar maximum stress produced brittle failure in HDPE (Fig. 5).

Although the development of this power to fracture concept is still in its fledgling state, the results are extremely promising. Brittle cracking in polyethylene is a result of the material being exposed to low stresses for long periods of time. Techniques which only measure energy associated with fracture ($J_{1C}$) neglect the rate of the fracture processes and thus are insufficient to distinguish differences in brittle crack propagation resistance between polyethylene resins. However, by incorporating both the time effect and the energy in a measure of the power to fracture, MDPE resins can be differentiated on the basis of their cracking resistance.
CONCLUSIONS

A technique to determine $J_{1C}$ from thin single edge notch specimens has been developed. Tests on HDPE showed that this new method results in $J_{1C}$ values which are equivalent to those obtained using the ASTM method. Using the new method brittle cracks were propagated in tough MDPE pipe resins, which did not crack in a brittle manner when tested using the monotonically loaded bending method (ASTM). Material differentiation with $J_{1C}$ was not possible because $J_{1C}$ neglects the rate of processes leading to fracture (time). Since brittle cracking in polyethylene pipes is a result of exposure to low stress for long periods of time, a method to measure the power to fracture, which incorporates both energy and time, has been developed and shown to be capable of differentiating MDPE pipe resins on the basis of their brittle cracking resistance.

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TABLE 4.1: RESULTS OF $J_{1C}$ FATIGUE TESTS

<table>
<thead>
<tr>
<th>Material</th>
<th>$J_{1C}$ (kJ/m²)</th>
<th>Power (GPa/sec)</th>
<th>$N_f-N_6$ (cycles)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MDPE’s</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ethylene/hexene 1</td>
<td>1.00</td>
<td>3.2</td>
<td>50,000</td>
</tr>
<tr>
<td>ethylene/butene</td>
<td>1.55</td>
<td>8.0</td>
<td>230,000</td>
</tr>
<tr>
<td>ethylene/hexene 2</td>
<td>1.63</td>
<td>6.8</td>
<td>150,000</td>
</tr>
<tr>
<td>ethylene/methylnpentene</td>
<td>1.70</td>
<td>4.5</td>
<td>110,000</td>
</tr>
<tr>
<td>HDPE</td>
<td>2.18</td>
<td>0.2</td>
<td>6,000</td>
</tr>
</tbody>
</table>

$N_f-N_6 = \text{number of cycles to fail minus number of cycles at a crack length of six millimeters}$
Figure 4.1) J-R curve for the ASTM SENB specimen. The solid curve is a power law fit to the filled squares, which are the data within the specified data window. The data window is confined by the dotted lines. The solid blunting line and dashed 0.2 mm blunting line offset are also shown. The insert is a schematic of a crack blunting mechanism.
Figure 4.2) Potential energy plotted as a function of crack length. The potential energy is proportional to the area above the loading curve in the hysteresis loop (insert).
Figure 4.3) Comparison of the ASTM and fatigue J-R curves. The solid blunting line and dashed 0.2mm blunting line offset are also shown.
\[ \sigma_{\text{max}} = 12.4 \text{ MPa} \]

Figure 4.4) Crack tip profile shortly before and immediately after initiation at a \( \sigma_{\text{max}} = 12.4 \) MPa. Massive blunting has occurred.
\( \sigma_{\text{max}} = 8.2 \text{ MPa} \)

Figure 4.5) Crack tip profile shortly before and immediately after initiation at a \( \sigma_{\text{max}} = 8.2 \text{ MPa} \). Minimal blunting has occurred. The crack profile has been highlighted.
Fatigue ($\sigma_{\text{max}} = 8.2\text{MPa}$)

ASTM E813

0.25 mm

Figure 4.6) Crack tip profiles of the fatigue SENT and the ASTM SENB specimen.
Figure 4.7) Schematic of damage growth and crack propagation mechanism.
Figure 4.8) Number of cycles as a function of crack length for four different fatigue conditions.
Figure 4.9) Potential energy as a function of crack length for four different fatigue conditions.
Figure 4.10) The power to fracture as a function of crack length for four different fatigue conditions.
Figure 4.11) Fracture surface of a MDPE tested at $\sigma_{\text{max}} = 6.7\text{MPa}$ showing the brittle to ductile transition near a ten millimeter crack length. The crack propagation direction is from left to right.
Figure 4.12) Results of power analysis for three additional MDPE's and a HDPE superimposed on figure 10.
CHAPTER 5:
PLANE STRAIN AND PLANE STRESS ANALYSIS OF
FATIGUE CRACK PROPAGATION IN
MEDIUM DENSITY POLYETHYLENE PIPE MATERIALS

ABSTRACT

Using the Crack Layer Theory, differences in damage formation under
different stress states during fatigue crack propagation in an ethylene-butene
copolymer were quantified and compared. Despite having vastly different
stress states and crack propagation behaviors, arc specimens (28 mm thick) and
SEN specimens (2 mm thick) were shown to have the same specific enthalpy of
damage, ~ 300 J/g, a parameter in the Crack Layer Theory which is a measure
of the material's intrinsic toughness. Damage in the SEN specimen consisted
of crazing and significant material yielding; the latter damage type is associated
with plane stress conditions. In the predominantly plane strain arc specimen,
material yielding was minimal compared to crazing, the dominant damage
form. After measuring these damage forms and applying the Crack Layer
Theory, the constancy of the specific enthalpy of damage was established.
Also, the dissipation coefficient, $\beta$, a second parameter of the Crack Layer
Theory, was shown to be a process-dependent parameter which was inversely
proportional to the lifetime of the specimen: $\beta_{\text{SEN}} = 4.6 \times 10^{-5}$, $\beta_{\text{arc}} = 1.1 \times 10^{-4}$, which corresponds to lifetimes of 140,000 and 30,000 cycles to failure,
respectively.

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INTRODUCTION

Many variables affect the fatigue crack propagation resistance of a polymer. For example, it has been found that introducing a compressive component in the fatigue cycle accelerates failure (1). Higher maximum stresses cause earlier and more ductile failures than do lower maximum stresses. Some polymers are sensitive to the type of loading waveform (2). Increasing the frequency may decrease the testing time, but it also increases the hysteretic heating of the specimen. Obviously, different polymers will have different fatigue crack propagation resistances, but the same polymer may also display different fatigue characteristics depending on the conditions under which it was processed. For example, if processing produces anisotropy within the polymer component, fatigue properties of the component will vary depending on orientation (3). The thickness of the specimen, which affects the stress state at the crack tip, is also an important factor which affects the fatigue crack propagation in a polymer. In these cases, the commonly used Paris plot (4) of the data will produce slopes and intercepts that are not only material dependent but also depend on the variables outlined above. This paper examines fatigue crack propagation behavior of an ethylene-butene copolymer under plane stress and plane strain conditions and employs the Crack Layer Theory (5) to identify a material dependent fracture resistance parameter.

EXPERIMENTAL

Extruded medium density ethylene/butene copolymer pipe, was provided by Philadelphia Electric Co. The pipe had an average outside diameter of 115 mm and a minimum wall thickness of 11 mm. Twenty eight millimeter wide arc-shape specimens with stainless steel reinforced six millimeter diameter loading holes placed 25 mm apart were prepared from the
pipe (Fig 1). A 2.5 mm deep razor notch was then introduced equidistant from each loading hole on the inner radius of each specimen (see e in Fig 1).

Single edge notched (SEN) specimens were prepared from the same pipe by milling strips from the pipe wall and then sanding the strips to a two millimeter thickness. The SEN specimens had final dimensions of 2 x 11 x 100 mm, and a 2.5 mm razor notch was introduced on the side which had been the inner bore of the pipe. Figure 2 shows the location of each geometry before it was cut from the pipe.

After the arc specimens were secured in a MTS servohydraulic testing machine with a pair of clevis grips, they were subject to sinusoidal tension-tension fatigue at 0.5 Hz, using a maximum load of 5.62 MPa (~25% $\sigma_y$). An R ratio ($\sigma_{\text{min}}/\sigma_{\text{max}}$) of 0.1 was used to minimize creep effects. An analog plotter was used to record hysteresis loops during the tests. A travelling optical microscope which was attached to a video camera, VCR, and monitor initially was used to measure and record the crack length. However, this method was inadequate due to crack tunneling at longer crack lengths, and a sharp graduated probe was inserted into the crack mouth to measure the extent of crack propagation during intermediate and late stages of crack growth. The SEN specimens were secured with standard flat grips and subject to the same loading conditions as the arc specimens. Due to the thinness of the SEN specimen, the crack length could be determined with the microscope, and the graduated probe was not necessary.

In order to determine the extent of damage associated with plane strain crack propagation, some tests were interrupted and the specimens were removed prior to failure. Sections to observe the damage zone surrounding the crack were prepared by using a low speed saw to cut the specimens in a
plane parallel to the crack propagation direction and orthogonal to the fracture surface (side view). Because of the high degree of pigmentation in the material, it was necessary to polish each section to a hundred micron thickness and apply a permanganate/acid etchant to elucidate the damage zone (6). After the completion of this procedure, transmitted light micrographs were used to determine the size and evolution of the damage zone.

The extent of plane stress damage (thinning) was determined by removing sections in a plane parallel to the crack front and normal to the fracture surface. The profile of these sections, the thickness reduction of the fracture surface, and the contraction observed in interrupted tests were all used to measure the extent of specimen thinning.

RESULTS

This study seeks to quantify differences in fatigue crack propagation between two very different specimen geometries. Although both geometries display some similar characteristics, the thick arc specimen is predominantly a plane strain geometry; the much thinner single edge notch geometry is predominantly a plane stress specimen. Because of these differences in stress states, these two geometries display very different fatigue crack propagation behavior. These differences will be quantified using the Crack Layer Theory.

The Crack Layer Theory considers the crack and its accompanying damage zone as a single entity which propagates through the material. This theory relates the crack propagation velocity \( \frac{da}{dN} \) to the energy release rate \( J \) and the irreversible work \( W_i \) by,

\[
\frac{da}{dN} = \frac{-\rho W_i}{\gamma \cdot R - J}
\]

where \( R \), the resistance moment, is a measure of the amount of damage around
the crack, β, the dissipation coefficient, is the fraction of irreversible work
(W_i) spent on fracture, and γ*, the specific enthalpy of damage, is the intrinsic
material resistance to crack propagation. The specific enthalpy of damage and
the dissipation coefficient are determined from the fit of the experimentally
determined da/dN, W_i, R, and J values.

Crack Velocity

The first of these values, da/dN, is determined from the first derivative
of the crack length (a) versus number of cycles (N) plot (Fig 3). This graph
shows that the crack in the SEN specimen takes over four times as long to
propagate to failure as does that in the arc specimen. Because of more easily
induced plastic flow at the crack tip, brittle cracking is typically slower in plane
stress specimens than in plane strain specimens. Subsequently, the
dependence of crack length on N is different for the two specimens. The
plateau observed near a nine millimeter crack length in the arc specimens’
crack propagation data (Fig. 3) is believed to be related to through the pipe
wall variances in morphology which were induced during processing, i.e., a
skin/core effect. This effect is not observed in the SEN specimen because of
its different geometry and origin from the pipe (Fig 2).

Figure 4 shows that the crack travels much faster in the arc specimen
than does the crack in the SEN specimen. From the slopes of the two curves, it
can also be noted that the crack accelerates faster in the arc than it does in the
SEN.

Irreversible Work

The irreversible work expended by the specimen per cycle is the area of
the load-displacement hysteresis loop (shaded area in Fig 5 insert) minus the
area of a loop recorded prior to crack initiation (accounts for bulk hysteresis).
Like the crack speed, there is a large difference in the values of the irreversible work for the arc and SEN geometries (Fig 5). Because the arc specimen is free to rotate around the loading pins, the resulting bending allows a larger grip displacement than that permitted with the SEN specimen. Also, the larger arc specimen naturally requires a larger load to obtain the same stress level as the SEN. The increased displacement and load produce a larger hysteresis loop and hence a larger irreversible work.

**Energy Release Rate**

The energy release rate, J, normalized to the initial specimen thickness, B₀, is the change in potential energy, P, with crack length,

\[ J = \frac{-dP}{da} B_0 \]  \[ (2) \]

In order to account for any crack advance which may occur during loading, the potential energy was taken to be the area above the unloading curve of the hysteresis loop. Thus, the change in this area per change in crack length is equal to the energy release rate. This method is similar to that outlined by Begley and Landes (7). Figure 7 shows the energy release rate plotted as a function of crack length. Since the crack propagates faster in the arc specimen than in the SEN, one would expect that the energy release rate would be greater for the arc geometry. This expectation agrees with the observed behavior.

Elastic-plastic energy release rate solutions only account for small scale yielding near the crack tip and thus neglect other forms of damage, such as extensive crazing and/or specimen thinning. In this system, it will be shown that these two types of damage are present in each specimen and can act as energy sinks. As a result, an elastic-plastic solution neglects a considerable
portion of the energy absorbing damage in these specimens, and therefore it should predict a larger energy release rate than that which is observed. A comparison of the Irwin corrected energy release rate \( G_{SEN} \) (8) for the SEN geometry to measured J values shows just such behavior (Fig 7).

**Resistance Moment**

To determine the resistance moment, the damage ahead of the crack tip must be identified and measured. During crack propagation in plane stress specimens, the crack tip is typically accompanied by a zone of uniformly yielded material. Previously (9,10), it has been shown that crack advance in the interior of the arc specimen (plane strain) is accompanied by a damage zone which consists of a main root craze which is partially enveloped by two smaller flanking side crazes. These crazes, which are essentially localized areas of extensive yielding and voiding, become larger at longer crack lengths and are located through the thickness of the specimen (Fig 8). However, the plane stress effect on the specimen edges induces yielding which masks the appearance of the interior crazes. Thus, the arc specimen, and as will be seen later, the SEN specimen, shows both plane stress and plane strain damage types, yielding and crazing, respectively. Since the resistance moment is the amount of damage ahead of the crack tip per unit crack extension, the contribution of each of these damage types will be computed separately and then added to determine the resistance moment.

**CRAZING**

The damage due to the crazes was calculated using the following equation,

\[
R_{craze} = \frac{V_f \sigma}{B_0 a}
\]  

[3],
where \( V \) is the volume occupied by the craze, \( f \) is the volume fraction of material within the craze, \( \delta \) is the material density \((0.9408 \text{ g/cc})\) \((11)\), \( B_0 \) is the initial specimen thickness, and \( a \) is the crack length. The volume of the craze zone is determined by optical micrography of a series of sections removed at mid-thickness from partially fractured specimens (Fig 8). For this analysis, it is assumed that the area of crazing observed at mid-thickness is constant throughout the specimen width. Microscopy of other sections, as well as sections and microscopy of other material, has shown that the extent of crazing decreases near the specimen edges; however, this decrease, and thus its effect on the resistance moment calculation, are minimal \((10,12)\). The volume fraction of material within the craze, \( f \), was determined to be 0.5 after extensive scanning electron microscopy of the craze structure \((3,9,10)\). The evolution of \( R_{\text{craze}} \) with crack length is shown in figure 9.

The crack in the interior of the SEN geometry is preceded by crazes which are similar to those observed in the arc specimen. However, the SEN root crazes are much smaller, and the side crazes, when visible, are very short (Fig 10). The resistance moment calculated from the volume of these crazes evolves in a manner similar to that observed in the arc specimen (Fig 9).

**YIELD ZONE**

Although the SEN geometry does display some plane strain characteristics (flat fracture surface, discontinuous crack growth, and crazing), because of its thinness \((2 \text{ mm})\) it has a significant plane stress contribution, especially when compared to the thick \((28 \text{ mm})\) arc specimen. The relative plane stress contribution can be gauged by the curvature of the arrest bands on the fracture surfaces of these two specimens (Fig 11). Whereas the SEN specimen has parabolic arrest bands, indicative of a plane stress component
through its entire thickness, the arrest bands on the arc specimen are straight for twenty-six out of its twenty-eight millimeter thickness. Since loaded plane stress polymer specimens typically demonstrate large scale yielding and material flow, this type of deformation should also be considered when calculating R. Figure 12 is a thinning (yielding) profile of the two geometries; the thickness (B) of the fracture surface has been normalized by the initial specimen thickness (B₀) and is plotted as a function of crack length. This graph illustrates that yielding is significant in the SEN geometry and minimal in the arc geometry. Since yielding acts as an energy sink just as crazing does, both damage types should be quantified in order to determine an accurate measure of the resistance moment.

During crack propagation, the yielded zone around the crack tip was observed to have an elliptical configuration (Fig 13). Thus, to quantify this zone, the evolution of its length, i.e., distance ahead of the crack tip, and width, i.e., distance above or below the crack plane, with crack length must be determined.

The zone width (zw) was determined by sectioning fractured specimens so that the profile of the thickness reduction could be observed (Fig 14 insert). The zone width was measured by determining the depth above or below the fracture surface at which thinning began. A plot of zone width versus crack length is shown in figure 14; the data fits a power law relationship,

\[ zw = 0.0086a^{1.86} \]  \hspace{1cm} [4].

Also shown in figure 14 is a plot of the zone width as predicted by the Von Mises yield criteria (13),

\[ \tau_y \quad (\theta=90) = \frac{5}{8\pi} \frac{K_1^2}{\sigma_y^2} \]  \hspace{1cm} [5].
where $K_1$ is the stress intensity factor. Since some deformation recovery is expected to happen immediately upon unloading (elastic), and since further time-dependent recovery (viscoelastic) will be accelerated by the sectioning process which creates local heating near the cutting plane, it was expected that the measured zone width would be less than that which is actually present during crack propagation. In lieu of an accurate way to measure zone width during crack propagation, the measured post-fracture zone width data was shifted to the theoretical zone width at the initial crack length, 2.5 mm, as determined by the Von Mises criteria, to account for the unmeasurable amount of deformation recovery. This adjustment for deformation relaxation resulted in the measured zone width data being multiplied by a shift factor of 6.2.

The zone length ($r_p$) was determined by fatiguing several specimens to different crack lengths. The partially fractured specimens were then removed from the MTS and freeze fractured after being placed in liquid nitrogen. Microscopy of the fracture surface revealed the location of the crack tip and the craze zone. The length of the yielded zone was measured by drawing a line parallel to the specimen edge from before the notch tip (Fig 15 insert). Because of yielding, the material has drawn away from this line, which represents the original specimen's edge. Using this method, the point beyond the crack tip at which the specimen returns to its original thickness was located. The distance from the crack tip to this point is the measured plastic zone length. Figure 15 shows that the plastic zone length grows exponentially with crack length,

$$r_p = 0.51 \exp(0.22a) \quad [6].$$

It is interesting to note that for shorter crack lengths the data are in the same
range as the plastic zone length predicted by Irwin (8),

$$r_p = \frac{k^2}{\pi \sigma_y^2} \quad [7]$$

The difference between the theoretical and measured behavior is slightly larger than the length of the craze (~0.5 mm) which precedes the crack tip. Thus, the measured data was multiplied by a factor, 0.53, to account for the contribution of the craze within the measured \( r_p \). This factor shifts the measured values of \( r_p \) to the theoretical \( r_p \) at the initial crack length (2.5 mm).

**CALCULATION OF RESISTANCE MOMENT**

Since the three dimensions of the yielded zone have been determined, it is now possible to calculate the volume of the plastic zone as the elliptical yield area seen on the specimen edge multiplied by the average thickness,

$$V = \frac{\pi zw r_p}{2}(t + t_o) \quad [8]$$

From the parabolic shape of the arrest bands in the SEN geometry (Fig 11a), it is deduced that this yielded zone extends through the entire two millimeter specimen thickness; curvature of the arrest bands is indicative of an edge (plane stress) effect. For the twenty-eight millimeter thick arc specimen the arrest bands only show curvature near the edges (Fig 11b). Approximately one millimeter from each edge the arrest bands become straight, indicating plane strain dominated fracture. Thus, the yielding contribution to fracture resistance in the arc specimen was only active in two out of the twenty-eight millimeter thickness. The volume of yielded material was then converted to a resistance moment by multiplying by its density (drawn material density = 0.9664 g/cc (11)) and dividing by the area of crack propagation,

$$R_{yield} = \frac{V\delta}{aB_o} \quad [9]$$
Since the craze zone is located within the yield zone its volume contribution should be removed from the $R_{\text{yield}}$ value. Because the $R_{\text{craze}}$ value was calculated using a volume fraction of 0.5 (half of the craze is occupied by voids), its value must be doubled and then subtracted from $R_{\text{yield}}$.

$$R_{\text{yield-craze}} = R_{\text{yield}} - 2R_{\text{craze}}$$  \[10\]

These calculations yield two $R$ values, one for crazing, $R_{\text{craze}}$, and one for yielding, $R_{\text{yield-craze}}$. Rather than trying to separate the proportion of energy which is expended on each process, it would be best to add the two $R$ values. In doing so, one must consider the intensity of damage in each zone and weight the corresponding $R$ accordingly. It has been noted that crazing is a very localized, intense form of damage that forms membranes of yielded material and voids. The volume fraction of material within the craze is about 0.5 (3,9,10). On the other hand, general crack tip yielding is a more subtle, diffuse type of damage. The measured density of the yielded material after a uniaxial tension test was 0.9664 g/cc, as compared to 0.9408 g/cc for the undeformed polyethylene (11). If the scaling factor for the $R$ values is based on these density changes, the yielding will only be 5.5% as effective as the crazing,

$$\frac{\Delta \delta_{\text{yield}}}{\Delta \delta_{\text{craze}}} = \frac{|0.9408 - 0.9664|}{|0.9408 - (0.5)(0.9664)|} = 0.0559$$  \[11\]

Using this factor the two resistance moments can be summed accordingly.

$$R = R_{\text{craze}} + 0.0559R_{\text{yield-craze}}$$  \[12\]

The plot of this combined resistance moment as a function of crack length is shown in figure 16. The combined resistance moment for the plane strain arc specimen, in which crazing was much more extensive, and in which the edge effects, $r_p$ and $zw$, were assumed to be the same as in the SEN, is also displayed
in figure 16.

Linearized Crack Layer Fit

Having established all of the variables of equation [1], it may now be written in the linear form,

\[
\frac{J}{R} = \gamma^* - \beta \frac{W_i}{R t_o da/dN}
\]  \[13\]

By plotting \(J/R\) against \(W_i/(R t_o da/dN)\), the constants \(\gamma^*\) and \(\beta\) can be determined from the \(y\)-intercept and slope, respectively (Fig 17). These plots show that \(\gamma^*\) is relatively constant for the two geometries, \(\gamma^*_{SEN} = 290\) J/g and \(\gamma^*_{arc} = 310\) J/g, indicating that the specific enthalpy of damage is dependent on the material and not the specimen geometry (Table I). On the other hand, \(\beta\), the dissipation coefficient, is larger for the arc specimen than for the SEN specimen, \(\beta_{SEN} = 4.6 \times 10^{-5}\) and \(\beta_{arc} = 1.1 \times 10^{-4}\). Since \(\beta\) is a process dependent parameter which represents the fraction of irreversible work expended on damage, it seems appropriate, that for the SEN specimen, a smaller \(\beta\) (less work expended on fracture processes) would increase the lifetime of the specimen (Table I).

From these results, it seems apparent that the Crack Layer Theory yields a parameter, \(\beta\), which ranks the fatigue lifetime, and that the specific enthalpy of damage, \(\gamma^*\), is relatively constant despite changes in stress state, crack plane orientation, and loading methods. Hence, \(\gamma^*\) can be considered to be a property of the material.

DISCUSSION

This work quantifies the effect of plane strain and plane stress on damage formation and crack propagation, and it uses the Crack Layer Theory to relate the differences caused by the varied stress states. The two specimens
compared are quite different. Although both specimens show signs of both stress states, i.e., yielding (plane stress) and crazing (plane strain), the SEN is predominantly a plane stress specimen, especially when compared to the much thicker arc specimen. Also, because the arrest bands on the arc fracture surface lack the curvature of those on the SEN fracture surface (Fig 11), it can be concluded that the arc specimen has a larger plane strain contribution.

Another difference between specimens which should be noted is that while the arc specimen is free to rotate about its loading pins (Fig 1), the SEN specimen was loaded using regular flat grips, thus subduing any specimen rotation. From figure 2, it should also be noted that these specimens have crack plane orientations which differ by 90°. The effect of each of these differences on the mechanism of crack propagation has been described in a previous paper (9).

The analysis outlined in the results section is relatively simple, with the exception of the resistance moment calculation. The energy release rate, irreversible, work, and crack velocity can be easily measured from the hysteresis loops and crack propagation data. However, the calculation of the resistance moment requires significant sectioning and microscopy of fractured and partially fractured specimens to quantify the size of the damage zone. In addition, it was necessary to make several theoretically sound assumptions when the direct measurement of a quantity was unattainable.

Specifically, correction factors were applied to the zone width and plastic zone length data. It is believed that the zone width correction factor accounts for unmeasured deformation recovery due to elastic and viscoelastic processes. The correction factor for the plastic zone length was meant to account for the craze at the crack tip which effectively increases the crack length and thus the stress intensity at the crack tip. As a result of this
artificially increased crack length the measured plastic zone length was greater than it would have been without the craze, which was measured and added to the resistance moment separately. In addition to these justifiable correction factors, an assumption exists that the relative effect of each damage type is dependent on the material density change in each. Intuitively, it seems clear that crazing, which is a very intense localized yielding phenomenon, would absorb more energy per volume than the more diffuse large scale yielding. Since this difference could not be measured directly, the relative changes in density were used as a scaling factor and assumed to be proportional to the effect of each damage type on the crack resistance of the specimen. Despite these limitations, this analysis is a solid first attempt to quantify different damage types and relate them to the observed fatigue crack propagation behavior.

The nearly identical values of the specific enthalpy of damage and the dissipation coefficients which correlate with the specimen lifetimes cannot be simply a fortuitous coincidence of this analysis. The various quantities in the Crack Layer Theory equation are dramatically different in the two cases. Furthermore, the extent of each type of damage is different in the two geometries. Thus, the obtaining of essentially the same value for the specific enthalpy of damage is indicative of a material parameter which is geometry independent.
CONCLUSION

The differences in fatigue crack propagation behavior between a plane stress and a plane strain specimen can be quantified and related via the Crack Layer Theory. Damage in the predominantly plane stress SEN specimen is a combination of crazing and material yielding, whereas yielding in the arc specimen is minimal compared to the amount of crazing. The dissipation coefficient ranks the fatigue life of the specimen, whereas the specific enthalpy of damage is a material constant independent of specimen geometry.

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Table 5.1: Comparison of SEN and Arc Specimens

<table>
<thead>
<tr>
<th>specimen</th>
<th>$\gamma^*$</th>
<th>$\beta$</th>
<th>$N_f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>SEN</td>
<td>290</td>
<td>$4.6 \times 10^{-5}$</td>
<td>140,000</td>
</tr>
<tr>
<td>arc</td>
<td>310</td>
<td>$1.1 \times 10^{-4}$</td>
<td>30,000</td>
</tr>
</tbody>
</table>

$\gamma^*$ - Specific Enthalpy of Damage
$\beta$ - Dissipation Coefficient
$N_f$ - Number of Cycles to Failure
Fig 5.1: Schematic diagram of the arc specimen (c), upper and lower clevis grips (a,b), steel reinforcing sleeves (d), and notch (e).
Fig 5.2: Respective origins of arc and SEN geometries.
Fig 5.3: Comparison of crack propagation kinetics of arc (+) and SEN (o) specimens.
Fig 5.4: Comparison of crack speed versus crack length for the arc (+) and SEN (o) specimens.
Fig 5.5: Irreversible work versus crack length behavior for both arc (+) and SEN (o) specimens. The irreversible work is proportional to the area of the hysteresis loop (shaded area of insert).
Fig 5.6: The difference between the areas above the unloading curves is proportional to the energy release rate.
Fig 5.7: Measured energy release rate for arc (+) and SEN (o) specimens, as well as the Irwin corrected elastic-plastic solution for SEN specimen.
Fig 5.8: Sequence of interrupted arc specimens in the brittle region. The vertical arrows mark the notch tip location.
Fig 5.9: Plot of resistance moment as calculated only from the volume of crazes at the crack tip for arc (+) and SEN (o) specimens.
Fig 5.10: Comparison of the craze zones in the arc and SEN geometries at a 3.5 mm crack length.
Fig 5.11a: Fracture surface of SEN specimen showing notch, brittle crack region, and ductile tearing region. The crack propagation direction is from left to right. Significant plane stress edge yielding can be observed.
Fig 5.11b: Fracture surface of arc specimen showing notch, brittle crack region, and ductile crack region. The crack propagation direction is from top to bottom. The edges of the specimen show trivial plane stress yielding.
Fig 5.12: The thickness of fracture surface normalized by the initial specimen thickness shows a great deal of thinning in the SEN (o) and trivial thinning in the arc (+) specimen.
Fig 5.13: Schematic illustration of the plane stress yield zone observed on the specimen edges.
Fig 5.14: Zone widths as measured from sections of a fractured SEN specimen plotted versus the crack length. The dashed line is the theoretical \( zw \) as determined by the Von Mises yield criteria.
Fig 5.15: Plot of measured plastic zone length versus crack length as compared to the Irwin plastic zone model (dashed line). The insert shows how \( r_p \) was measured from freeze fractured interrupted SEN experiments.
Fig 5.16: Resistance moment, including effect of thinning and crazing, for the arc (+) and SEN (o) specimens.
\[ \frac{J}{R} = \gamma^* - \frac{\delta W_i}{R t_0 \frac{da}{dN}} \]

Fig 5.17a: Linearized crack layer plot for SEN specimen.
Fig 5.17b: Linearized crack layer plot for arc specimen.
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