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Investigating the existence of a threshold stress intensity for slow crack growth in high-density polyethylene

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The existence of a threshold stress intensity for failure by slow crack growth under plane strain conditions for single-edge notched tensile specimens and for three-point bending was investigated at 42° C. It was found that the threshold for the nucleation of damage was between 0.074 and 0.063 MPa $m^{1/2}$ and the threshold for complete failure was about 0.08 MPa $m^{1/2}$.

1. Introduction

The concept of a critical parameter for crack initiation such as K_c or J_c is usually applicable for a fracture process that is independent of time. In the case of fracture that involves an incubation period and then slow crack growth to produce the ultimate fracture, the concept of threshold conditions, below which ultimate failure will never occur, is more complex. Because the rate of slow crack growth was found to vary as $K^{4.7}$, the determination of a threshold may simply depend on the minimum crack growth that can be measured with existing experimental techniques and the time limitations of the investigation.

There are very few direct experimental investigations to determine threshold conditions for failure in polymers by slow crack growth, even though the matter has important technological implications as pointed out by Kausch [1] who gives no data on the subject. Dean and McCartney [2] give a theory for the existence of a "threshold level of stress intensity below which crack growth under a static load should not occur". In trying to verify the theory on PVC they experienced experimental difficulties with the collection of stable crack growth data. Part of their difficulty was caused by the complexity of having a combination of plane stress and plane strain fracture. Döll [3] measured the rate of crack growth by fatigue for PMMA over a wide range of K. He found that the slope of log $\mathrm{d}a/\mathrm{d}N$ against log K was finite down to the lowest value of $\mathrm{d}a/\mathrm{d}N$ that he measured; this result did not indicate the existence of a threshold in the stress intensity for PMMA under fatigue loading. Hertzberg and Manson [4] give plots of $\mathrm{d}a/\mathrm{d}N$ against K for many polymers and show that

$$\mathrm{d}a/\mathrm{d}N \approx K^m \quad (1)$$

down to the lowest values of K. These results indicate that for fatigue loading there is no threshold stress intensity below which $\mathrm{d}a/\mathrm{d}N$ becomes zero. However, in the case of unnotched specimens, Hertzberg and Manson [4] show that many polymers exhibit a fatigue limit in the conventional S–N plot where we are reminded that the existence of a fatigue limit may depend on the fact that practically all tests are terminated before about 10^7 cycles. These results suggest that there may not be a threshold for the crack growth rate if there is a pre-existing defect. The observed fatigue limit in the unnotched specimens may be associated with a threshold for the initiation of crack growth.

This investigation concerning the existence of threshold conditions for failure by slow crack growth under a constant stress was prompted by the extensive data on the life time of PE gas pipes and the long-standing concern about the durability of PE insulation for electrical wires. Note that this investigation is concerned with a constant applied stress as contrasted with constant strain conditions where stress relaxation conditions could lead to a threshold simply because the stress may relax to a very low level as proposed by Dean and McCartney [2]. During the past 7 years our extensive research programme has focused on the phenomenon of slow crack growth in many kinds of PE. Generally the conditions of stress, initial notch depth, temperature, and method of loading have been chosen so that test results could be obtained in the minimum time. If the stress is too high, however, the failure mode is ductile. If the stress, notch depth and temperature are too low, the rate of damage is too small to be measured within the time frame of the experiments which has ranged from 10 min to more than 1 y, depending on the resin. In this investigation on the possible existence of a threshold, it is necessary to explore the very lowest stress level and smallest possible notch depths. Thus, the time frame of the experiment must be extended as much as possible. It was decided to limit the duration of any test to 10 w. If a threshold cannot be measured within this time

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frame, then it is at least expected that an upper bound can be established.

The polyethylene resin that was chosen is the same resin on which extensive slow crack growth data had been obtained by Brown and co-workers [5–8] where the stress intensities were generally above 0.2 MPa $m^{1/2}$. The temperature of 42° C was chosen because the most extensive data base was available at this temperature and because it was sufficiently close to room temperature where the results of the investigation are most useful. Being above room temperature, the kinetics of the process are significantly faster because the activation energy is 100 kJ mol^{-1}. All experiments were conducted under plane strain conditions.

Lu and Brown [5, 6] found that the initial rate of damage in pre-notched specimens essentially determined the course of the subsequent fracture process and the time to failure. The initial rate of damage as measured by the change in crack opening displacement (COD) with time obeyed the following equation

$$\dot{\delta}_0 = C\, e^{-Q/RT} \sigma^m a_0^n \quad (2)$$

where C is a constant that depends on the method of loading and the type of resin and its thermal history. Q is the activation energy and equals 100 kJ mol^{-1} for linear homopolymers of PE and is independent of molecular weight as determined by Huang and Brown [7]. σ is stress and a_0 is notch depth, $m = 5$ and $n = 1.9$. All the data that determined Equation 2 also satisfied the following equation

$$\dot{\delta}_0 = A\, K^{4.6} \quad (3)$$

even though $n \neq m/2$. Thus Equation 3 is a more useful but less accurate representation of the damage process when compared to Equation 2. Further work by Bassani *et al.* [8] showed that Equation 3 could be improved by using the J-integral instead of K. Lu and Brown [5, 6] showed that the time to failure can be predicted from $\dot{\delta}_0$ where

$$t_f = \frac{\alpha\, a_0 + \delta_c}{\dot{\delta}_0} \quad (4)$$

where $\alpha = 10°$ is the apex angle of the triangular zone of damage, δ_c is the COD at the initiation of crack growth. This equation was useful in the cases where t_f but not $\dot{\delta}_0$ could be measured.

The plan of the research was to explore the thresholds in σ and a_0 below which measurements of the slow crack growth process were no longer possible within the time frame and the measuring limitations of the experiment. Thus, when σ or a_0 was held constant the other was reduced as much as possible.

It was found that crack initiation could be observed at a value of K as low as 0.074 MPa $m^{1/2}$. At a value of K of 0.063 MPa $m^{1/2}$ no evidence of damage could be observed by a microscopic observation. These results indicate that a threshold stress intensity for slow crack growth exists between 0.078 and 0.063 MPa $m^{1/2}$ for this material.

A theoretical calculation for the threshold based on the assumption that crack growth cannot occur unless yielding occurs at the root of the initial notch gives a value which is consistent with the experimental observations.

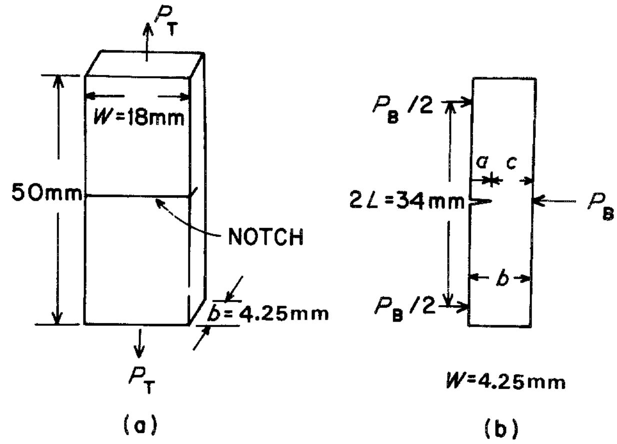

Figure 1 Geometry of specimens (a) single-edge notch tension; (b) three-point bending.

2. Experimental details

The resin was the same Marlex 6006 used in previous investigations [5–8]: $M_n = 19\,600$; $M_w = 130\,000$. The compression-moulded plaques were 4.3 mm thick. Some materials were slowly cooled and others were quenched. The geometry of the single-edge notch tension specimen and the three-point bending specimen are shown in Fig. 1. Each specimen was notched very slowly with a fresh razor blade so that the notching did not produce an observable zone of damage at the root of the notch. The geometry of the razor blade is shown in Fig. 2. The three-point bending (TPB) specimens consisted of slowly cooled material under a constant stress of 12 MPa and the notch depth was varied from 400 to 11 μm. The single-edge notch tensile specimen (SENT) consisted of quenched material that was subsequently annealed at 80° C for 5 d to relax the residual stresses. The notch depth was constant at 0.4 mm whereas the stress was varied from 1 to 10 MPa. The temperature for all tests was 42° C.

The COD was measured at the root of the notch with a filar eye piece on the microscope. The minimum measurable value of the COD was about 2 μm. For

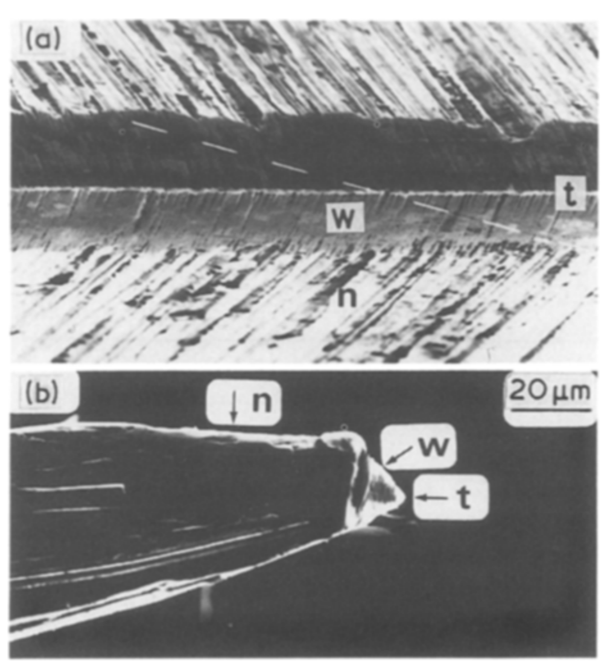

Figure 2 Geometry of a razor blade: (a) top view; (b) cross-section.

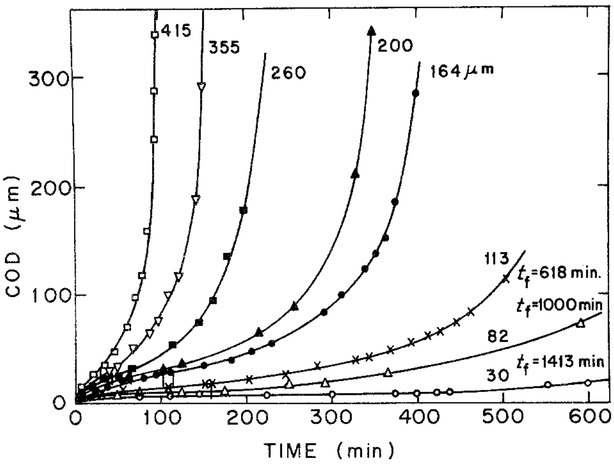

Figure 3 COD plotted against time for three-point bending, slowly cooled. 42° C, 12 MPa and various notch depths.

three-point bending, when the notch depth was below 30 μm and for single-edge notch tension when the stress was below 5 MPa, the initial COD rate was too small to be measured. In these cases only the time to failure was measured.

The general experimental approach was to determine the minimum value of stress and notch depth for which experimental data could be obtained. The new data at low stress intensities were also correlated with the extensive data that had been obtained by Lu and co-workers [5, 6, 8] at higher values of $K > 0.2\,\text{MPa}\,\text{m}^{1/2}$.

3. Results

3.1. Effect of notch depth

The results in this section are directed towards the determination of the minimum notch depth that will produce slow crack growth. Fig. 3 shows the notch opening plotted against time for various notch depths for three-point bending and 12 MPa stress. The stress was chosen to be as high as possible and still to produce brittle fracture. When the stress becomes much greater than one-half the yield point, ductile failure is produced. The beginning of fibril fracture corresponds to the time when the curves begin to accelerate. The slope of the curve prior to fibril fracture is related to the time to failure in accordance with Equation 4. For notch depths below about 30 μm, the

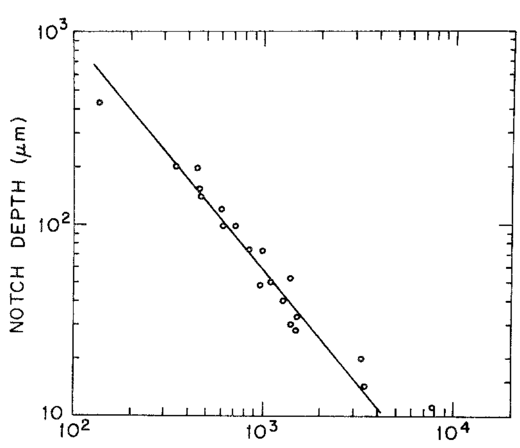

Figure 4 Notch depth plotted against time to failure, for three-point bending, σ = 12 MPa. $t_f = 2.6 \times 10^3 a_0^{-0.8}$ (min, a_0 μm).

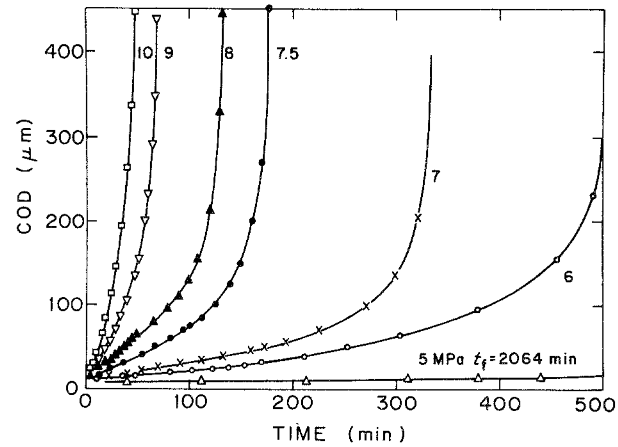

Figure 5 COD plotted against time for SENT, quenched, 42° C, a_0 = 400 μm and various stresses.

notch opening was not measurable and only the time to failure could be reliably measured. The notch depth against time to failure is plotted in Fig. 4 from which

$$t_f = 2.6 \times 10^3 a_0^{-0.8} \text{ (min, } a_0 \text{ in } \mu\text{m)} \qquad (5)$$

The one point that is displaced appreciably from the curve is for a_0 = 11 μm. The measurement of a_0 becomes unreliable when the notch depth is about 10 μm. The unreliability stems from the uncertainties of the surface roughness and the surface distortion produced by the razor blade. Fig. 4 shows that for a_0 greater than 14 μm the relationship between t_f and a_0 is consistent and in agreement with all the previous data by Lu and co-workers [5, 6, 8]. The departure of the 11 μm point from the curve suggests a threshold effect, but the uncertainty in the measurement of a_0 makes this indication very unreliable.

3.2. Effect of stress

The experiments in this section consisted in keeping a_0 constant and determining the minimum stress at which crack growth could be observed. Fig. 5 shows the notch opening plotted against time for various stresses and a_0 = 400 μm for single-edge notched tensile specimens. The stress was converted to stress intensity where

$$K = Y \sigma a_0^{1/2} \qquad (6)$$

Y, for a_0 = 400 μm and the specimen geometry of Fig. 1a, is equal to 2.1 (Paris and Sih [9]). The values of Y for three-point bending depend on the ratio of notch depth to specimen width [9]. The stress intensity is plotted against time to failure in Fig. 6. Data for K

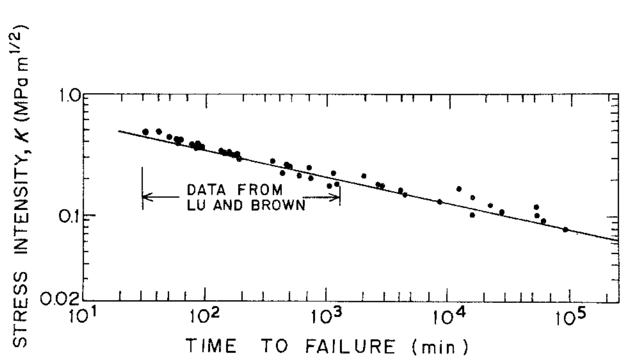

Figure 6 Time to failure plotted against K: a_0 = constant; SENT specimen, 42° C. $t_f = 0.98K^{-4.6}$ (min).

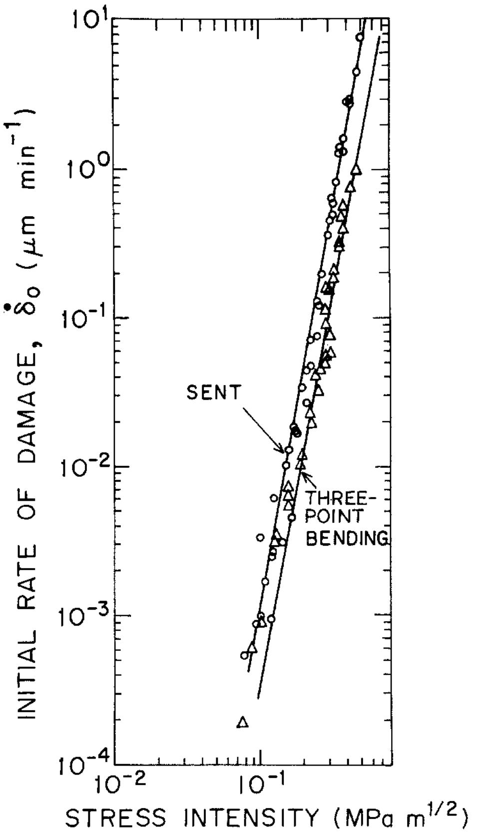

Figure 7 Initial rate of damage plotted against stress intensity for SENT quenched and three-point bending, slowly cooled material.

greater than 0.2 MPa m$^{1/2}$ are taken from the previous work by Lu and co-workers [5, 8]. The new data for K down to $K = 0.078$ MPa m$^{1/2}$ follows the same curve as that for the higher values of K where

$$t_f = 0.98\,K^{-4.6} \text{ (min, } K \text{ in MPa m}^{1/2}) \quad (7)$$

For $K = 0.078$ MPa m$^{1/2}$, the time to failure was 10^5 min. Thus, it can be said that if a threshold for slow crack growth exists under the conditions of single-edge notch tension at 42° C, then the threshold value of K is less than 0.078 MPa m$^{1/2}$.

All the data for slow crack growth at 42° C for Marlex 6006 as produced by this investigation and the previous investigations by Lu and co-workers [5, 6, 8] are brought together in Fig. 7 where the initial rate of damage as measured by the initial slope of the COD against time curve is plotted against K. In those cases where only t_f could be measured, the initial rate of damage was calculated from Equation 4. Fig. 7 shows that the single-edge notch data are separate from the three-point bending data. It has been shown by Bassani *et al.* [8] that using the J integral instead of K brings the two curves closer together; however, there is still a difference because the materials have a different thermal history. From the viewpoint of whether a threshold for K exists, the data indicate that for K ranging from 0.50 to about 0.078 MPa m$^{1/2}$ the same process occurs. If there is a threshold for K, it is below 0.078 MPa m$^{1/2}$.

3.3. Microscopic evidence for a threshold

Another way to investigate the existence of a threshold for slow crack growth is to look for microscopic evidence for crack initiation. If the conditions are such that no evidence of damage is detected, then it can be stated that those conditions represent a threshold that depends on the resolution of the microscopic technique that is used. The technique involves slicing the specimen and viewing the interior with the SEM. The specimen is held in jig which slightly opens the notch in order to make the damage more evident. The usual form of damage is shown in Fig. 8a at low magnification. Fig. 8b, at a high magnification, shows the tip of the damaged zone where the damage is nucleated. The very first evidence for damage at the root of the initial notch consists of the microcrazes shown in Fig. 8b. The resolution of the technique is about 0.1 μm for observing the thickness of a craze whose length to thickness ratio is about 10.

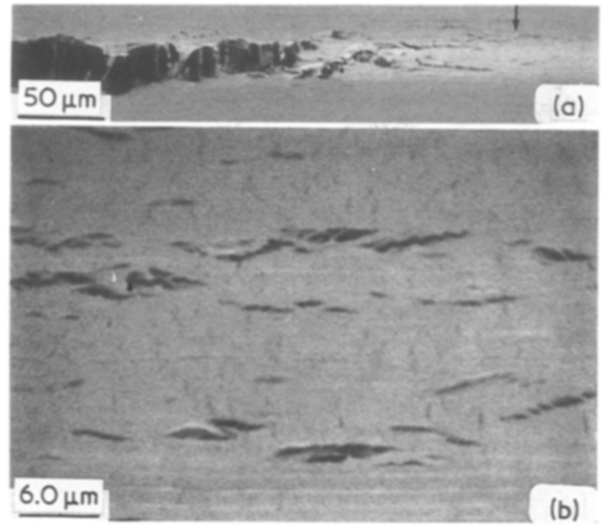

Figure 8 Typical damaged zone: (a) cracked region plus process zone; (b) enlarged view of the process zone indicated by the arrow.

Microscopic investigations of the root of the notch prior to loading do not show these microcrazes. The single-edge notch tensile specimen, after being exposed to $K = 0.074$ MPa m$^{1/2}$ for 5.2×10^4 min exhibited the microcraze shown in Fig. 9. Whether 0.074 MPa m$^{1/2}$ would produce failure after 10^5 min is not known. Other specimens which were exposed to 0.063 MPa m$^{1/2}$ for 5×10^4 min did not show any evidence of a microcraze. Thus, it can be stated that for this particular microscopic technique a threshold for craze nucleation exists between 0.074 and 0.063 MPa m$^{1/2}$. This value is slightly smaller than the value of 0.078 MPa m$^{1/2}$ under which complete failure was observed.

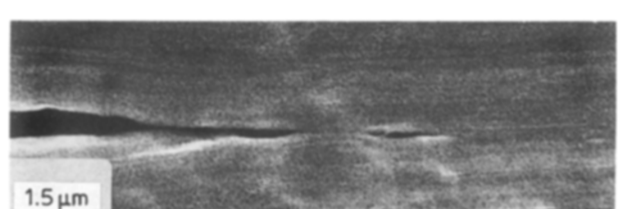

Figure 9 Very early stage in the formation of the process zone, 1.75 MPa, $a_0 = 0.4$ mm, $K = 0.074$ MPa m$^{1/2}$, 42° C, loading time = 5.2×10^4 min.

3.4. Theoretical calculations of a threshold

The concept of K is based on the assumption of an infinitely sharp notch, so that even at the lowest applied stress, the local stress should be sufficient to produce some form of damage by yielding or crazing. In examining the conditions for a threshold for the nucleation of damage it is necessary to know the actual geometry of the notch at the microscopic level and to realize that real notches have a finite sharpness. Fig. 2 shows the geometry of the razor blade where the tip of the razor blade has a radius of curvature of about 2 μm. The radius of curvature of the notch produced by this razor blade has not been measured but it is greater than 2 μm and is associated with a wedged portion beyond the tip. The effective notch radius is less than about 9 μm.

The local stress at the root of an elliptical notch is given by Inglis [10]

$$\sigma_{\text{local}} = \sigma\left[1 + 2\left(\frac{a}{r}\right)^{1/2}\right] \qquad (8)$$

where σ is the applied stress, a is one-half of the long axis of the ellipse and r is the radius of curvature of the tip. If the local stress does not exceed the yield point of the material, then no damage can be produced. Thus a condition for a threshold is

$$\sigma[1 + 2(a/r)^{1/2}] < \sigma_y$$

For the case where $2(a/r)^{1/2} \gg 1$ and using the above expression, the condition for a threshold stress intensity is given by

$$K < \frac{yr^{1/2}\sigma_y}{2} \qquad (9)$$

If $r = 9\,\mu$m; $Y = 2.1$; $\sigma_y = 20$ MPa at 42° C, then $K < 0.06$ MPa m$^{1/2}$ is the threshold condition for the nucleation of damage. This value is close to the value suggested by the microscopic observation and depends on the estimated value of r. In an unnotched specimen, r would depend on the size and shape of the inherent defects in the material. This threshold value should decrease with increasing temperature in the same way as does the yield point.

This threshold for nucleation as given in Equation 9 is time dependent in the same way that σ_y depends on time. Yielding in PE under a constant stress, occurs when the creep strain reaches a critical value, ε_y. Because the creep curves have the form [11]

$$\varepsilon = A\,\sigma^p\,t^q \qquad (10)$$

then the stress at which yielding occurs, σ_y, is given by

$$\sigma_y = \left(\frac{\varepsilon_y}{At^q}\right)^{1/p} \qquad (11)$$

Inserting Equation 11 into Equation 9 will give the time dependence for the threshold stress intensity for nucleating the damage zone.

Predictions of the size of the damage zone by the Dugdale theory have given good agreement with the experimental observations by Lu and Brown [5, 6] and Bhattacharya and Brown [12] for K_{th} values greater than 0.2 MPa. The Dugdale theory will predict a threshold value of K_{th} for the smallest value of the damaged zone that can be resolved by our microscopic technique where

$$K_{th} = (\Delta a)^{1/2}\,\sigma_y\left(\frac{8}{\pi}\right)^{1/2}$$

Δa is the smallest length of damaged zone that can be detected with our microscopic technique and is estimated to be about 1 μm. Thus, for $\sigma_y = 20$ MPa, $K_{th} = 0.03$ MPa m$^{1/2}$. This value is below the experimental limit of 0.063 MPa m$^{1/2}$. The Dugdale prediction does not work at low values of K because it is based on the assumption of an extremely sharp notch.

If the worst inherent defect in an unnotched specimen consisted of a circular hole, then the maximum stress is independent of the radius of the hole and has the value of 3. On this basis there would be a threshold stress of $1/3\,\sigma_y$ or 6.7 MPa. Experiments show that an unnotched specimen under a stress of 6.7 MPa would not fail within the time limitations of our experiments.

4. Discussion

Decreasing the notch and keeping the stress constant shows that if a threshold intensity exists it is below 0.087 MPa m$^{1/2}$ (Fig. 4). For an 11 μm deep notch with $K = 0.078$ MPa m$^{1/2}$, the observed time for failure was twice as great as the value obtained by extrapolation of the data from higher K values. Possibly some change in the mechanism is taking place for $K \approx 0.078$ MPa m$^{1/2}$, but this point is somewhat uncertain because the measurement of the notch depth is unreliable at about 10 μm.

Decreasing the stress and keeping the notch depth constant also indicates that if a threshold stress intensity exists it is below 0.078 MPa m$^{1/2}$ in agreement with the variable notch tests.

Microscopic examinations of damage with the SEM showed that damage could be observed after 5.2×10^4 min exposure to 0.074 MPa m$^{1/2}$ but could not be observed at 0.063 MPa m$^{1/2}$ after the same time. Thus, it is concluded that the threshold for the nucleation of damage is between 0.074 and 0.063 MPa m$^{1/2}$. It is of interest that for 42° C and the time limitation of these experiments, the thresholds for time to failure and craze nucleation are about the same. The mechanism for nucleating damage is different than that required for crack growth and subsequent failure. The nucleating process consists in converting virgin material into a craze. In addition, crack growth requires the disentanglement of the fibrils and further nucleation of the craze at its tip during growth. Without the nucleation of a craze, fracture should never occur.

5. Conclusion

It is concluded that for single-edge notched tensile specimens and for three-point bending at 42° C for Marlex 6006, the threshold stress intensity for producing failure by slow crack growth under plane strain conditions is about 0.08 MPa m$^{1/2}$.

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