JOURNAL OF MATERIALS SCIENCE 19 (1984) 413–422

The fracture toughness of tough polyethylenes by a novel high pressure technique

R. W. TRUSS*, R. A. DUCKETT, I. M. WARD
Department of Physics, The University of Leeds, Leeds, UK

The failure of two grades of polyethylene has been studied using the technique of torsion under superposed hydrostatic pressure. The behaviour of unnotched samples of both grades was ductile at all pressures and strain-rates. However, at sufficiently high pressures both grades of polyethylene (including a tough copolymer) failed in a brittle manner when a surface notch was exposed to a suitable pressure fluid. Measurement of fracture stresses for various notch depths lead to a value for a critical stress intensity factor at each of several pressures. A linear extrapolation was then used to estimate the critical stress intensity factor at atmospheric pressure to be 1.11 ± 0.05 MN $m^{-3/2}$ and 1.68 ± 0.08 MN $m^{-3/2}$ for the homopolymer and copolymer, respectively. An independent measurement at atmospheric pressure for the homopolymer using compact tension geometry yielded a value of 1.28 ± 0.02 MN $m^{-3/2}$ confirming the accuracy of the extrapolation procedure and that the effect of the environment on the behaviour was not substantial.

1. Introduction

Tough grades of polyethylene are finding increasing utilization in gas- and water-pipe applications. Limited fracture data is available for many of these important industrial polymers since they behave in a ductile fashion under normal laboratory test conditions. Additionally, many polyethylenes are known to fail in an unexpectedly brittle fashion after long times under loads well below their yield stress especially when in contact with certain environments such as detergents [1]. Laboratory tests to simulate these types of failure require experiments to run over an excessively long period of time. It would thus be exceedingly useful to develop short-term tests to obtain fracture data on these tough polyethylenes.

We recently reported a technique of applying hydrostatic pressure to a tough gas-pipe grade polyethylene to induce brittle failure in this material in a short-term laboratory test [2]. This technique was based on the fact that for most polymers, including polyethylene, the pressure dependence of the yield stress is greater than that of the fracture stress and so with increasing hydrostatic pressure, the polymer can be made to undergo a ductile to brittle transition as the yield stress exceeds the fracture stress. The fracture stress in this previous work was a somewhat arbitrary figure in that the tests were conducted on notched specimens and the fracture stress would vary with notch depth. In this present work, fracture data have been obtained in terms of the critical stress intensity factor, or fracture toughness, K_c, which is a material parameter independent of specimen geometry. Two grades of polyethylene have been tested in torsion under hydrostatic pressures sufficiently high that they failed in a brittle fashion. K_c for both materials was determined as a function of pressure and these results have been extrapolated to atmospheric pressure to give a value of the fracture toughness for a tough gas-pipe grade polyethylene at atmospheric pressure.

*Present address: I.C.I. Australia, Research Department, Newsom Stress, Ascot Vale, Melbourne, Australia.

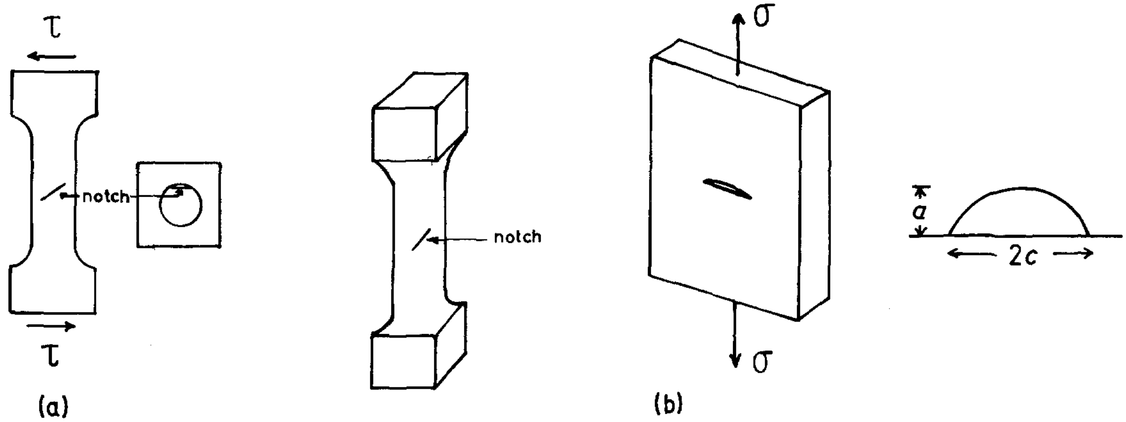

Figure 1 (a) The torsion specimen containing a notch at 45° to the specimen axis. (b) The equivalent surface notch in relation to the tensile component of the shear stress.

2. Theory

In this work, torsion tests under superposed hydrostatic pressure were conducted on notched solid cylindrical specimens. The specimen geometry is shown in Fig. 1a. The specimens were notched at 45° to the axis of the specimen so that the shear stress, τ, in the surface of the cylinder, due to the torque on the specimen was equivalent to a tensile stress, σ, of equal magnitude perpendicular to the face of the crack. As a first approximation, the notch in the cylindrical specimen subjected to torsion was modelled as a semi-elliptical surface crack in a sheet in tension (Fig. 1b). Irwin [3] has analysed this type of crack and has given the stress intensity factor, K, as

$$K^2 = \frac{1.2\pi\sigma^2 a}{\Phi^2 - 0.212(\sigma/\sigma_y)^2} \tag{1}$$

where σ_y is the yield stress and Φ is an elliptical integral given by

$$\Phi = \int_0^{\pi/2} (1 - k^2 \sin^2 b)^{1/2} db$$

where $k^2 = 1 - (a/c)^2$ where a and c are the minor and major axes of the ellipse. Irwin obtained Equation 1 by solving the mathematical problem of an elliptical crack in an infinite solid and applying to it corrections for a free surface normal to the crack through the major axis of the ellipse and for plastic yielding effects near the crack edge. From Equation 1, it can be seen that by plotting the square of the fracture stress, σ_F^2, against Q/a where $Q = \Phi^2 - 0.212\ (\sigma_F/\sigma_y)^2$, a straight line should be obtained from whose slope the fracture toughness, K_c, can be calculated.

It was observed in previous work [2] that the fracture stress of notched specimens of polyethylene increased approximately linearly with pressure, P, so that

$$\sigma_F = \sigma_0 + \alpha P$$

where α is a constant and σ_0 is the fracture stress at atmospheric pressure. The factor, Q, would be expected to vary non-linearly with pressure since both the yield stress and the fracture stress of a specimen with a given notch are increased with pressure but at different rates. However, since Φ^2 has an order of magnitude of 1 and the fracture stress would be expected to be at least half the yield stress, Q cannot vary by more than approximately 15% and can be considered, as a first approximation, to be a constant with pressure. Thus,

$$\begin{aligned} K_c &= \sigma_F \left(\frac{1.2\pi a}{Q}\right)^{1/2} \\ &= \sigma_0 \left(\frac{1.2\pi a}{Q}\right)^{1/2} + \alpha P \left(\frac{1.2\pi a}{Q}\right)^{1/2} \\ &= K_c(0) + \alpha' P \end{aligned} \tag{2}$$

where $K_c(0)$ is the fracture toughness at atmospheric pressure and α' is a constant.

It is probable that Equation 2 is not completely general in that the linear dependence of σ_F on pressure may not hold at very high hydrostatic pressures. Chan and Williams [4] have reported a non-linear dependence of K_c on temperature for polyethylene in the region of the γ relaxation and we have reported changes in the yield behaviour of polyethylene which can be associated with the mechanical relaxations of the polymer [5]. The γ relaxation which normally occurs at $\sim -100^\circ$ C at atmospheric pressure would be moved to higher temperatures with increasing hydrostatic pressure but since the mechanical relaxations are moved to higher temperatures at a rate of $\sim 15^\circ$ C per $100\,\mathrm{MN\,m^{-2}}$ hydrostatic pressure, no non-linearity in the pressure dependence of K_c due to the γ relaxation would be expected up to the limit of hydrostatic pressure used in this work (i.e. $700\,\mathrm{MN\,m^{-2}}$).

Equation 2 suggests that by conducting tests to determine the fracture toughness of polyethylene at different levels of hydrostatic pressure where the pressure is high enough so that materials which are ductile at atmospheric pressure fail in a brittle fashion, the results can be extrapolated to give a value of the fracture toughness at atmospheric pressure. The validity of this technique was checked by applying it to a relatively brittle low molecular weight linear polyethylene for which K_c at atmospheric pressure could be determined by an independent technique using compact tension specimens. The procedure was then used on a high toughness gas-pipe grade polyethylene copolymer to obtain a value of its fracture toughness at atmospheric pressure.

3. Materials and procedure

Two grades of polyethylene were tested in this work and their characterizations are listed in Table I. Rigidex 50 was a relatively low molecular weight linear polyethylene while Rigidex 002 40 was a gas-pipe grade material and consequently contained a yellow dye for identification purposes.

High pressure torsion specimens of Rigidex 50 were injection moulded to the shape shown in Fig. 1a and had diameters of 6, 8 and 10 mm with a gauge length of 25.4 mm. Specimens of Rigidex 002 40 were machined from compression-moulded blocks and had diameters of 10 mm and gauge lengths of 31.8 mm. The specimens were notched in a special jig which permitted a new razor blade to be pressed to variable depths into the gauge length at 45° to the axis of the specimen. The actual dimensions of the notch were measured under a microscope after the specimens had been fractured. The specimens were enclosed by a thin rubber sheath into which an environment of 10 vol% of Igepal in de-ionized water was introduced. Igepal is a nonyl phenoxypoly (ethyleneoxy) ethanol which is a nonionic surfactant recommended as a standard environment for environmental stress cracking tests in polyethylene [6].

The high pressure torsion apparatus which has been described previously [7] allowed tests to be conducted up to a pressure of $700\,\mathrm{MN\,m^{-2}}$. All the tests were conducted at 20° C (± 0.5° C) at a strain rate of $9.5 \times 10^{-4}\,\mathrm{sec^{-1}}$.

The basic data were in the form of torque–twist curves which were analysed using the Nadai equation to calculate the shear stress–strain data [7].

An independent measure of K_c for Rigidex 50 at atmospheric pressure was obtained using compact tension specimens cut from slow-cooled compression-moulded sheets of thickness 4.3 mm. In the compact tension technique a stable crack was propagated across the specimen which was pulled apart on the Instron at a constant cross-head speed of $0.05\,\mathrm{cm\,min^{-1}}$. The load was measured at given crack lengths from which the stress intensity factor was calculated using the Srawley and Gross boundary collocation technique [8].

4. Results and discussion

Fig. 2 shows the stress–strain curves for notched specimens of Rigidex 50 tested in Igepal solution at various levels of hydrostatic pressure up to $700\,\mathrm{MN\,m^{-2}}$ at 20° C and a strain rate of $9.5 \times 10^{-4}\,\mathrm{sec^{-1}}$. For a given notch depth, the strain to failure decreased and the fracture stress increased with increasing pressure. Since the stress–strain curves did not show a maximum which could be considered as the yield stress, a failure strain of 5% was taken below which the curves were considered as brittle failure. In previous work [2, 5], a 2% offset stress has proved a reasonable measure of the yield stress and the 2% offset stress was

TABLE I

Grade	$\bar{M}_w$	$\bar{M}_n$	Density	Branch content
Rigidex 50	101 450	6 180	0.972	linear
Rigidex 002 40 (Batch no. 6109)	134 000	20 800	0.945	~ 4 butyl groups/ 10^3 C atoms

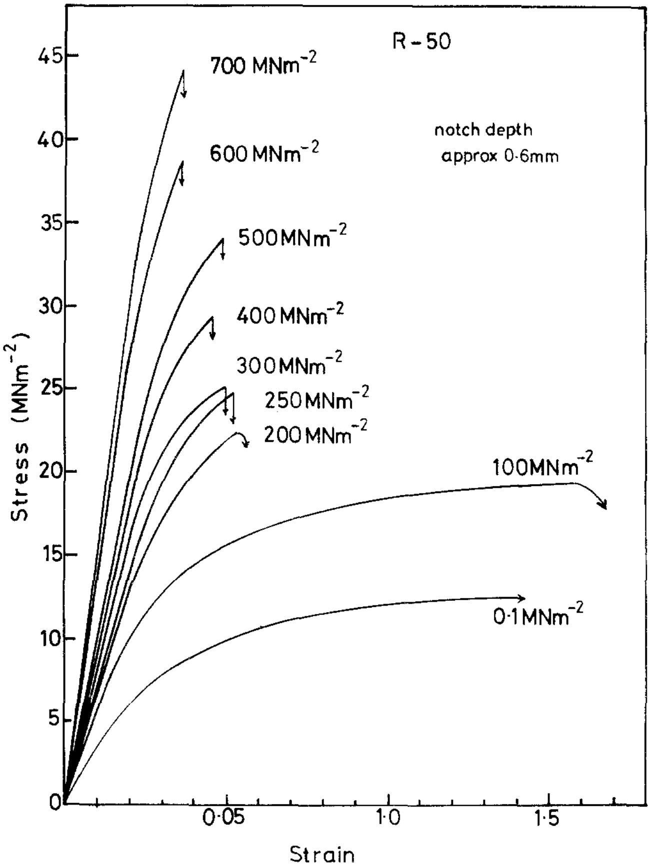

Figure 2 Shear stress–strain curves for Rigidex 50 at the pressures marked. Temperature 20° C, strain rate 9.5×10^{-4} sec^{-1}, environment 10% Igepal in water, notch depth 0.6 mm.

reached at strains greater than 5% for all pressures used in this work. At hydrostatic pressures below 250 MN m^{-2}, Rigidex 50 failed only after a degree of ductility and for these cases, the fracture mechanics approach could not be applied. At pressures higher than 250 MN m^{-2}, Rigidex 50 failed at strains less than 5% for all but the most shallow notches and showed a fracture strain which decreased with increasing notch depth. Specimens which failed at less than 5% strain were analysed using Equation 1.

Fig. 3 shows plots of σ_F^2 against Q/a for Rigidex 50 tested at different hydrostatic pressures from 250 to 700 MN m^{-2}. The lines plotted in Fig. 3 are the best fit straight lines through the origin from whose slope the fracture toughness, K_c, was calculated. Fracture tests at a pressure of 400 MN m^{-2} were conducted on 6, 8 and 10 mm diameter samples. No significant difference was observed in the results obtained on specimens of different diameter. K_c was plotted as a function of pressure for Rigidex 50 in Fig. 4. The error bars in Fig. 4 are ± one standard error of the mean value of K_c obtained at each pressure in Fig. 3. The best fit line through the high pressure data points for Rigidex 50 was extrapolated to atmospheric pressure to give a value of K_c at atmospheric pressure of 1.11 (± 0.05) MN m$^{-3/2}$.

The value of K_c for Rigidex 50 obtained from compact tension is 1.28 ± 0.02 MN m$^{-3/2}$, only slightly higher than the value of 1.11 ± 0.05 NM m$^{-3/2}$ obtained from the extrapolation of the high-pressure data. The closeness of these values suggests that the extrapolation procedure does result in realistic lower-bound estimates for K_c at atmospheric pressure. The small difference between the compact tension results (obtained in air) and the torsion data (obtained in Igepal solution) is, however, more than three times larger than the standard error and is therefore considered to be significant. The fact that the value of K_c in the presence of the environment was only some 15% lower than in air suggests that Igepal only affects the fracture behaviour to a marginal extent at room temperature on these time scales.

Fig. 5 shows stress–strain curves for notched specimens of Rigidex 002 40 tested in Igepal solution at various levels of hydrostatic pressure up to 700 MN m^{-2} at 20° C and a strain rate of

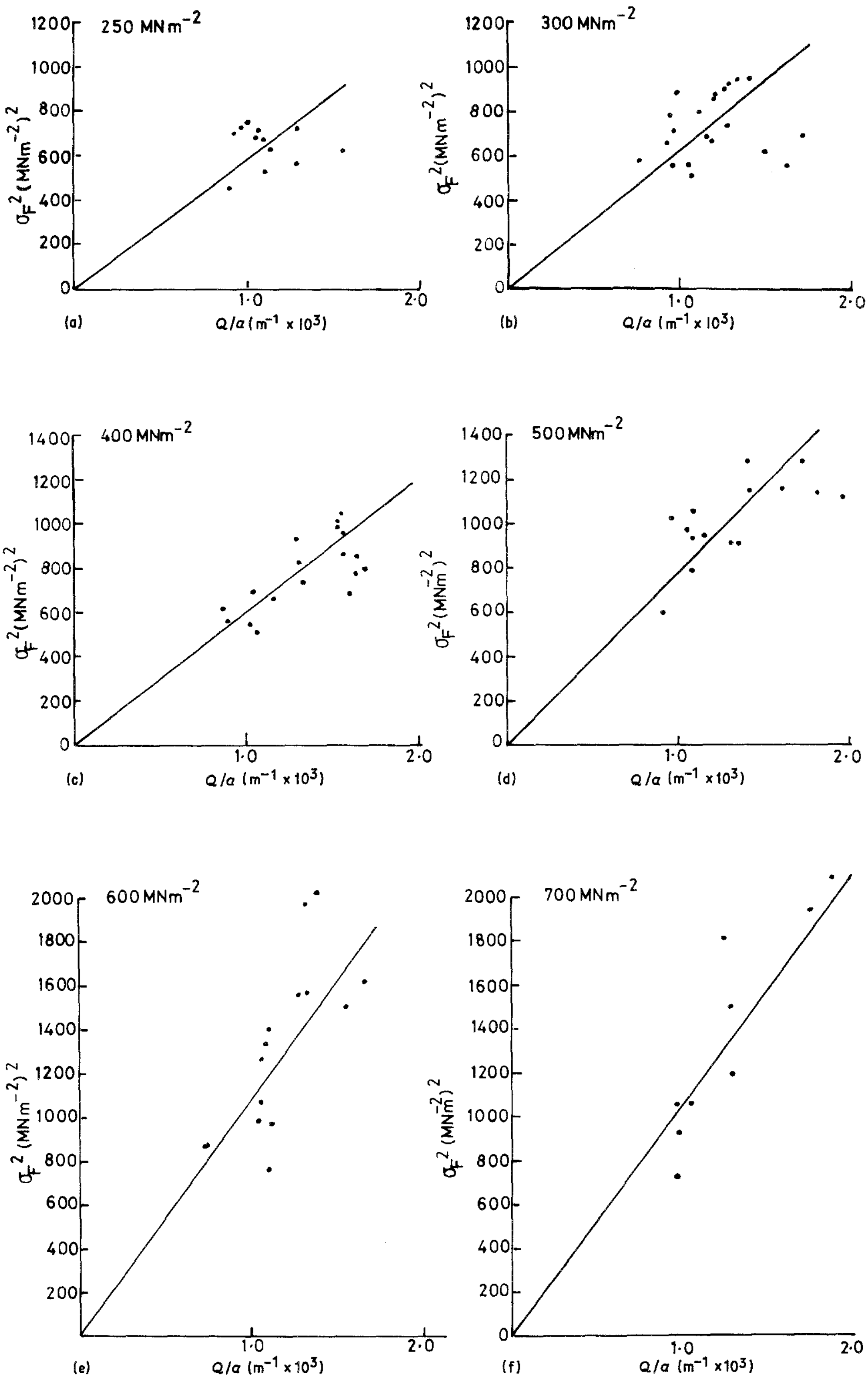

Figure 3 Plots of σ_F^2 against Q/a for Rigidex 50 at the pressures marked. Conditions identical to those for Fig. 2. See text for definition of Q.

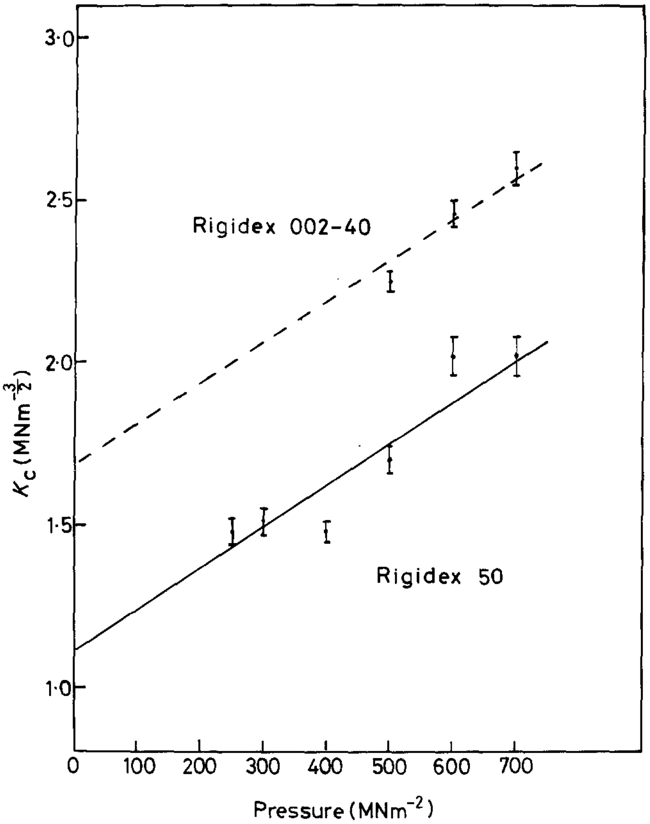

Figure 4 Plot of fracture toughness against pressure for Rigidex 50 (full line) and Rigidex 002-40 (dotted line). Data from Figs. 3 and 6.

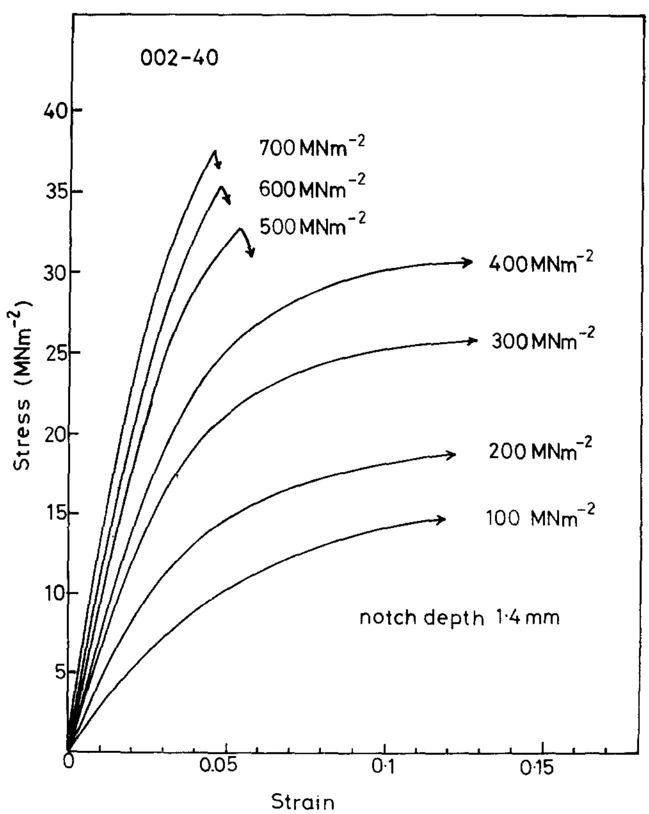

Figure 5 Shear stress–strain curves for Rigidex 002-40 at the pressures marked. Conditions identical to those for Fig. 2. Notch depth 1.4 mm.

$9.5 \times 10^{-4}\ \mathrm{sec}^{-1}$. Considerably higher pressures than those required for Rigidex 50 were necessary to induce brittle failure in Rigidex 002 40 and fracture data could only be obtained at pressures above $500\ \mathrm{MN\,m^{-2}}$. Fig. 6 shows plots of σ_F^2 against Q/a for Rigidex 002 40 at hydrostatic pressures between 500 and $700\ \mathrm{MN\,m^{-2}}$. The value of K_c at each pressure was again calculated from the slope of the best fit straight line through the data points and is plotted against pressure in Fig. 4. Since data for Rigidex 002-40 could only be obtained over a narrow range of pressure, an accurate measure of the pressure dependence of K_c could not be determined. Previous work [2] has shown that the strain-rate behaviour of the fracture stresses of Rigidex 50 and 002-40 are similar and that there is an overall equivalence between pressure and strain rate. It was therefore assumed that the pressure dependence of K_c of Rigidex 002-40 is the same as that for Rigidex 50 and this results in a predicted value of $1.68 \pm 0.08\ \mathrm{MN\,m^{-3/2}}$ for Rigidex 002-40 at atmospheric pressure.

Chan and Williams [4] have reported measurements of K_c at atmospheric pressure as a function of temperature for a number of polyethylenes. They gave a value of K_c of $\sim 1.25\ \mathrm{MN\,m^{-3/2}}$ for Rigidex 006-60 at 20° C. This is a linear polyethylene with a slightly higher molecular weight than Rigidex 50 and the values of K_c are quite comparable. Chan and Williams were unable to obtain a value of Rigidex 002-40 at 20° C but gave a value of $\sim 6.4\ \mathrm{MN\,m^{-3/2}}$ for -120° C. They also indicated from estimated values that K_c decreased only marginally with increasing temperature between -120 and -60° C. If such a trend continued to 20° C the value of K_c for Rigidex 002-40 would be $\sim 6.0\ \mathrm{MN\,m^{-3/2}}$, which must be compared with the value of $1.68\ \mathrm{MN\,m^{-3/2}}$ measured here. It is surprising that Chan and Williams indicated little change in K_c of Rigidex 002-40 in the range -120 to -60° C since the two other grades of polyethylene tested by them showed a marked decrease in this region which they linked to the γ-relaxation. If a similar decrease occurred in Rigidex 002-40, the values of K_c for Rigidex 002-40 found in this work would seem reasonable, especially if it is considered that the value obtained here may be as much as 15% lower than the value in air due to the presence of the Igepal environment.

Roberts et al. [9] measured for fracture toughness of a medium density polyethylene (Hostalen GM 5050, density 0.954, also intended for gas-pipe application) at high rates over a range of temperatures. At low temperatures the fracture toughness became approximately temperature independent with a value of approximately 2.7 to $3.2\ \mathrm{MN\,m^{-3/2}}$, in general agreement with the values quoted in this investigation.

During the progress of this work a second batch of Rigidex 002-40 became available (batch no. 6910) which was significantly tougher than the batch tested here. Injection-moulded samples of this material were tested but they could not be made to fail in a brittle fashion at pressures up to $700\ \mathrm{MN\,m^{-2}}$. Some specimens with fairly severe notches did fail at just over 5% strain at $700\ \mathrm{MN\,m^{-2}}$ pressure, giving a K_c of approximately $3\ \mathrm{MN\,m^{-3/2}}$. This would suggest a value of K_c at atmospheric pressure of approximately $2.1\ \mathrm{MN\,m^{-3/2}}$ but a facility to test at pressures higher than $700\ \mathrm{MN\,m^{-2}}$ would be required to state these values with confidence.

It is instructive to use the data presented in this work to calculate the radius of the plane strain plastic zone [10]

$$r_p = \frac{1}{4(2)^{1/2}\pi}\left(\frac{K_c}{\sigma_t}\right)^2$$

and the minimum specimen thickness for valid plane strain fracture toughness measurements [11],

$$t_{\min} = 2.5\left(\frac{K_c}{\sigma_t}\right)^2.$$

Direct measurements of the shear yield stress σ_y were available at each pressure and these were converted into equivalent tensile yield stresses σ_t assuming a von Mises yield criterion modified to allow for the observed linear dependence on hydrostatic pressure. These values of σ_t were then combined with directly measured values of K_c and the calculated values of r_p and $t_{\min}$ are shown in Table II. They reveal that the plastic zone size in Rigidex 002-40 is substantial at atmospheric pressure and confirm the need to use very large specimens in order to obtain valid plane strain fracture toughness data. It is also apparent that the specimen thickness used for the compact tension results on Rigidex 50 in this work (4.3 mm) does not strictly satisfy the minimum thickness requirements and are therefore most likely to overestimate the true plane strain fracture toughness).

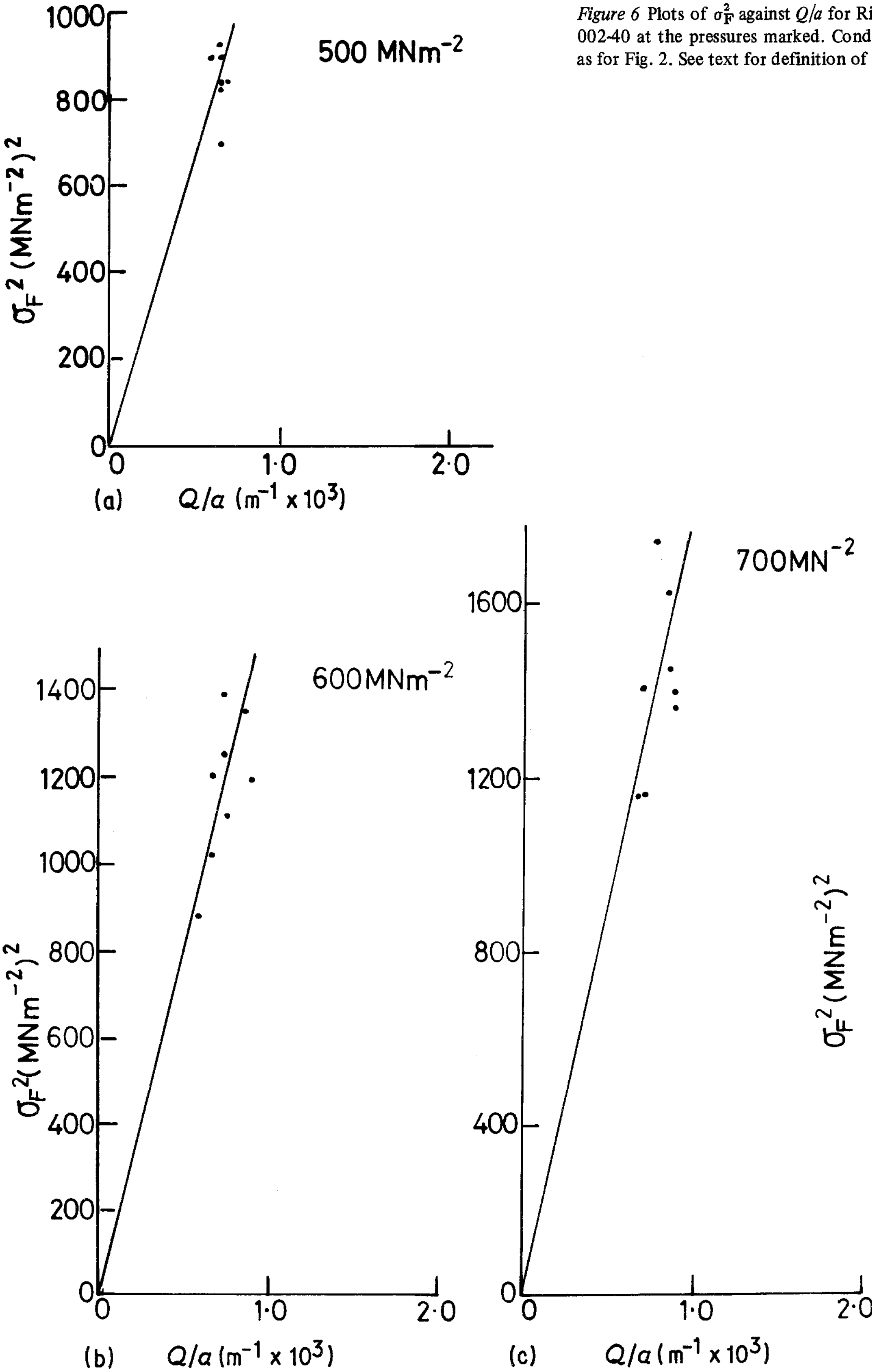

Figure 6 Plots of σ_F^2 against Q/a for Rigidex 002-40 at the pressures marked. Conditions as for Fig. 2. See text for definition of Q.

TABLE II Plane strain plastic zone sizes [10] and minimum specimen thicknesses [10] required for valid plane strain fracture toughness testing

	Rigidex 50					Rigidex 002-40				
P(MN m^{-2})	σ_y(MN m^{-2})	σ_t(MN m^{-2})	K_c(MN m$^{-3/2}$)	r_p(μm)	t_{min}(mm)	σ_y(MN m^{-2})	σ_t(MN m^{-2})	K_c(MN m$^{3/2}$)	r_p(μm)	t_{min}(mm)
0.01	10	16.1	1.11	252	11.9	6.5	10.8	1.68	1284	60.5
100	15.6	25.9	1.29	130	6.2	10.6	17.6	1.81	561	24.4
300	28.2	46.8	1.49	54	2.5	21.2	35.3	2.06	181	8.5
500	41	68	1.75	35	1.7	32.4	53.9	2.31	97	4.6
700	53.6	88.9	2.0	27	1.3	43.4	72.2	2.56	67	3.14

The plastic zone sizes are reduced substantially at high pressures and confirm the feasibility of obtaining valid plane strain fracture toughness data even with specimens of 10 mm diameter as used in this study.

5. Conclusions

A reasonable estimate of the fracture toughness, K_c, at atmospheric pressure of tough grades of polyethylene which are normally ductile at atmospheric pressure can be determined by evaluating K_c as a function of pressure at high pressures where these materials behave in a brittle fashion. These data can then be extrapolated to atmospheric pressure. This technique when applied to a relatively brittle polymer, Rigidex 50, gave a value of K_c at 20° C and a strain rate of 9.5×10^{-4} sec^{-1} at atmospheric pressure of 1.11 (± 0.05) MN m$^{-3/2}$. This value was slightly lower than the value of K_c obtained for this material in compact tension tests of 1.28 (± 0.02) MN m$^{-3/2}$ but this difference can possibly be attributed to the presence of an Igepal environment in the high pressure tests. K_c was also found as a function of pressure for a tough gas-pipe grade polyethylene, Rigidex 002-40. When these data were extrapolated to atmospheric pressure, a predicted value of K_c for Rigidex 002-40 at 20° C and a strain rate of 9.5×10^{-4} sec^{-1} of 1.68 (± 0.08) MN m$^{-3/2}$ was obtained.

Acknowledgements

We are grateful to Dr D. Sims, PERME, Waltham Abbey, for injection moulding specimens, and to BP Chemicals Ltd., for provision of compression-moulded blocks of Rigidex 002-40.

References

1. J. B. HOWARD, in "Engineering Design for Plastics", edited by E. Baer (Van Nostrand, New York, 1964) Ch. 11.
2. R. W. TRUSS. R. A. DUCKETT and I. M. WARD, *J. Mater. Sci.* **16** (1981) 1689.
3. G. R. IRWIN, *J. Appl. Mech.* **29** (1962) 651.
4. M. K. V. CHAN and J. G. WILLIAMS, *Polymer Eng. Sci.* **21** (1981) 1019.
5. R. W. TRUSS, P. D. CLARKE, R. A. DUCKETT and I. M. WARD, *J. Polym. Sci.: Polym. Phys. Ed.* in press.
6. ASTM D2522, Annual Book of ASTM Standards, Part 36 (1980).
7. S. H. JOSEPH and R. A. DUCKETT, *Polymer* **19** (1978) 837.
8. J. E. SRAWLEY and B. GROSS, NASA Report TN D-3820 (1967).
9. D. R. ROBERTS, J. F. MANDELL and F. J. McGARRY, Research Report R81-3 Massachusetts Institute of Technology, April (1981).
10. F. A. McCLINTOCK and G. R. IRWIN, in ASTM STP381 (1965) p. 85.
11. ASTM Designation E399 (1981) Part 10 Annual Standards.

Received 20 December 1982
and accepted 19 May 1983