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Precise evaluation of fast fracture velocities in acrylic polymers at the slow-to-fast transition

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Transient fast fracture velocities at the onset of the slow-to-fast transition in polymethylmethacrylate (PMMA) have been measured precisely in single-edge-notched specimens of various geometries by using ultrasonic fractography. Little sign of the fracture velocity of the order of 10^0 m sec^{-1} have been detected on the instability onset. The fast fracture starts from a point source almost instantaneously. The initial velocity has been shown to fall in a small range, 90 to 150 m sec^{-1}, almost independent of the loading speeds from 0.1 to 100 mm min^{-1} and specimen temperature from − 50 to 40° C, with exceptional cases for specimens loaded slowly (0.1 mm min^{-1}) at a low temperature (− 50° C). As the final minimum velocity of an arresting crack, a value of 42 m sec^{-1} has been obtained under room temperature. Crack propagation in low molecular weight PMMA has been shown to be more temperature, as well as loading rate, dependent than in higher molecular weight PMMA.

1. Introduction

Fracture velocity measurements provide information on the mechanical and chemical phenomena controlling the fracture process. Various methods have been used for the measurements, including electric methods, schlieren optical methods and high speed photography. As far as the fast fracture velocity change at such highly transient stages as the slow-to-fast instability onset or crack arrest in glassy polymers is concerned, no precise measurements have been performed due to technical difficulties stated below, although the values are of importance, because they are closely related to the mechanisms involved in the transient phenomena [1, 2].

For fast cracks in glassy polymers, particularly in polymethylmethacrylate (PMMA) [3–8], an electric method employing conductive lines on a specimen surface has conveniently been used by many investigators for its simplicity. An electric impedance method has also been employed using high frequency [9] or d.c. [10] current through a conductor-coated specimen surface. Opto-electric methods have been applied to the fracture velocity record of PMMA, and transient velocity changes during fast propagation have been studied [11]. However, these electric and opto-electric methods, or even other reliable methods such as high speed photography, can hardly assess such highly transient stages as are dealt with here; the slow-to-fast instability onset in glassy polymers involves rapid fracture velocity change of more than three figures, and fast cracks are suddenly arrested without leaving any evidence of a slow crack region on fractured surfaces. The difficulty with the conventional methods is associated with technical problems arising from, firstly, synchronization of measuring devices with the transient phenomena whose initiation is scattered widely in time as well as in location. Insufficiency of the measurement precision inherent in these methods should become another cause. Furthermore, application of these

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TABLE I Size of specimens tested

Configuration	Notch	l, w, t (mm)	a_{00} (mm)	PMMA ($M_v = 2.3 \times 10^6$)	PMMA (P(MMA/MA)) ($M_v = 2 \times 10^5$)	Temperature
Compact tension	Pre-cracked	50, 52, 5	~16	○		room temp.
Tensile	Pre-cracked	150, 35, 5	~ 8	○	○	−50 ~ 40° C
Tensile	V-shape	150, 35, 5	~ 8	○		room temp.
Tensile	2 mm-hole	150, 35, 5	~ 8	○		room temp.
Bend	Pre-cracked	60, 35, 5 (w) (h)	~ 8	○		room temp.
Charpy	Pre-cracked	60, 15, 5 35	6 ~ 8	○		room temp.

a_{00}: initial notch length, l: length, w: width, t: thickness, h: height.

methods has been restricted solely to one-dimensional crack propagation which should not always be the case for the transient cracks.

Ultrasonic fractography, originally developed by Kerkhof [12], seemed to be a successful method to overcome these drawbacks of the conventional methods, because it is basically thoroughly free from the synchronization problem, and available for two- or three-dimensional crack propagation with higher accuracy. Although some attempts to apply the Kerkhof method to polymer fracture, especially in PMMA, were reported previously [13–16], its application to glassy polymers involved substantial difficulties [17]. Recently, however, the present authors have experimentally confirmed that, by adopting suitable experimental conditions, this method is applicable to such glassy polymers as PMMA and epoxy for obtaining two-dimensional precise fast fracture velocities at transient stages [17]. In the present study, fast fracture velocities in acrylic polymers at the slow-to-fast instability onset, as well as crack arrest, are successfully evaluated by this method.

2. Experimental

Ultrasonic modulation tests were performed for two commercially available acrylic polymers of different molecular weights. One, formed by casting, had a higher molecular weight (M_v) of 2.3×10^6. The other formed by extrusion, was a random copolymer of a large content of methylmethacrylate and of a small content of methylacrylate, and its M_v was about 2×10^5. In order to distinguish this from the former, the notation P(MMA/MA) shall be used in the following. For the former, the specimen geometry of various types, including compact tension (CT), single edge notched (SEN) tensile, bend and Charpy types, were adopted, whereas for the latter only SEN-type specimens were tested. In all types of specimen geometry pre-cracking was made on one edge of the specimens by chisel impact onto a sawcut. For specimens of the SEN-type, the chisel cracking was further followed by fatigue cracking which employed a vibration frequency as high as 60 or 120 Hz. This procedure resulted in well-finished pre-cracks with a desired length which had a single crazed layer ahead of them. Table I summarizes the size of tested specimens, whose geometry is shown in Fig. 1. The initial pre-crack length in SEN plates was about one-fourth of the specimen width. Uniaxial loading of SEN, CT and bend specimens was performed in an Instron-type machine at a low and a high cross-head speed, 0.1 and 100 mm min^{-1}, respectively. The position of the ultrasonic transducer mounted on an edge

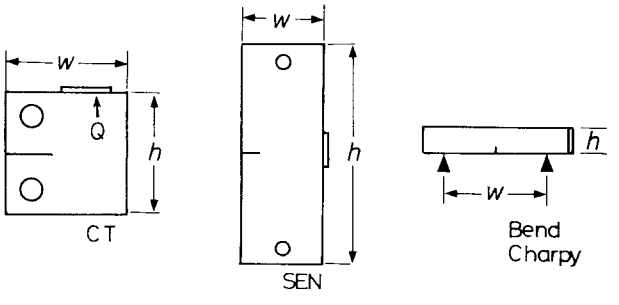

Figure 1 Specimen geometry and the position of the ultrasonic transducer, Q.

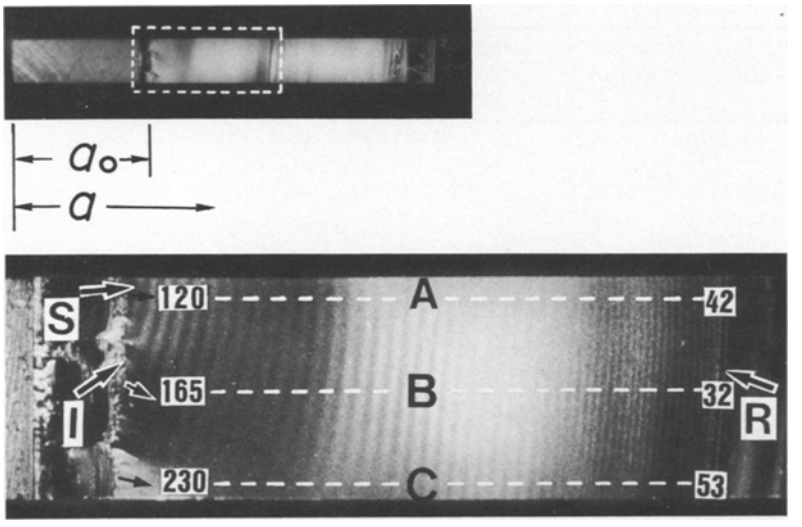

Figure 2 Ultrasonically modulated fracture surface of PMMA compact tension specimen.

of each specimen is illustrated in Fig. 1. Charpy specimens had the transducer on the top of each specimen. In order to maintain the weight balance on the Charpy-machine specimen holders, a counter balance was attached on another side of the specimen. A wider range of ultrasonic test frequencies from 200 kHz to 7.2 MHz was made available for the present test, and frequencies up to 900 kHz were mostly used. For SEN specimens the temperature was varied from -50 to 40° C by using a temperature bath attached in the tensile machine. The bath was equipped with a liquid nitrogen gas inlet for cooling down to -190° C and a fan heater for heating up to 90° C. In all tests, in order to avoid specimen heating through ultrasonic loading, care was taken so that the loading time did not exceed a few seconds. The fracture velocity, v_f, was evaluated from the wavelength of ultrasonic lines [12, 17] which was obtained with an optical microscope on a fracture surface.

3. Results and discussion

3.1. Compact tension tests for PMMA specimens

Fast fracture in CT specimens has often both transient stages, instability onset and crack arrest. Fig. 2 presents an example of the fracture surface of a PMMA CT specimen, which was fractured under a cross-head speed of 100 mm min^{-1}. The marks I and R show each location of the instability and the arrest line, respectively. On the fracture surface, a number of lines traversing in the direction of the specimen thickness are ultrasonic lines. Each ultrasonic line in this case presents the momentary position of the fast crack front during propagation. The location of the first ultrasonic line (see the mark S) as a matter of course gives the exact position where the instability took place. In all specimens throughout the present tests, the instability started from a point source and propagated radially, or two-dimensionally, in the initial course. It should be noted that no observable dense array of lines exists near the point S, which indicates that the transition from the slow-to-fast propagation occurred almost discontinuously, as is the case with SEN specimens [17]. The wavelength between the first and the second line is 310 μm, which gives an initial fracture velocity of 120 m sec^{-1} for an employed frequency of 410 kHz. This means that the fast fracture propagated stably already when it travelled a length of about 300 μm from the instability initiation point. The spatial resolution can be raised with a higher modulation frequency; using 860 kHz [17] and 3.6 MHz [15] waves, this figure has been shown to be shortened to 100 μm and 50 μm, respectively. Because the initial crack propagation from the instability point has a two-dimensional nature, the real crack speed along the instability line should differ considerably from position to position. In contrast to the initial value of v_f (v_{fc}) determined at the point S, the values obtained at the middle of, and at the opposite side of, the instability line are 165 and 230 m sec^{-1}, respectively (see Fig. 2). This means that if we had used the conducting line method and had measured the value of v_{fc} on the surface opposite to the crack initiation, an error of about 100% would have resulted, even if the syn-

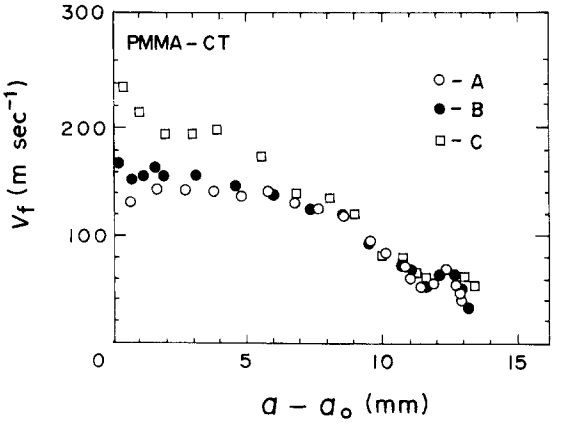

Figure 3 Change of the fracture velocity, v_f, along different paths A, B and C, on the fracture surface of the compact tension specimen shown in Fig. 2.

chronization problem had been dealt with successfully, and also the v_f measurement had been performed very precisely. The result shown in Fig. 2, therefore, indicates that when we refer to the "initial velocity" just after the instability its value must be related to the exact fast crack initiation location on the instability line. Likewise, the velocity change in the propagation process should also be referred to the definite path on a fracture surface along which the measurement is performed. In Fig. 3 is given the change of v_f measured along the three different paths, A, B and C, each defined on a fracture surface shown in Fig. 2. The abscissa $a - a_0$ denotes the fast crack length measured from the instability line. The length of a_0, the crack length at the onset of instability, was 16.5 mm in this case. Fracture velocity was measured in a direction perpendicular to the ultrasonic line, as is indicated by each arrow in Fig. 2. For the same value of $(a - a_0)$, a considerable velocity difference exists among the three paths up to the first half of the fast fracture area. The propagation became almost one-dimensional at the length of 6 to 7 mm, i.e. 120 to 140% of the specimen thickness. The fast crack was suddenly arrested after propagation of another 6 to 7 mm. In contrast to the instability onset, the arrest took place almost one-dimensionally. The final velocities obtained for the paths, A, B and C, were 42, 32 and 53 m sec^{-1}, respectively. The average velocity was 42 m sec^{-1}. It should be noted that the value of final velocity detected is far less than that of v_{fc}. Results of Figs. 2 and 3 elucidate that the present method is very advantageous for the two-dimensional fast fracture velocity measurement. In the following, the v_f measurement was performed along such a path as A in Fig. 2, which starts from the exact instability point.

3.2. Tensile tests for single edge notched PMMA specimens

In Fig. 4 are presented results for the v_f measurement for pre-cracked PMMA tensile specimens which were uniaxially loaded under temperatures

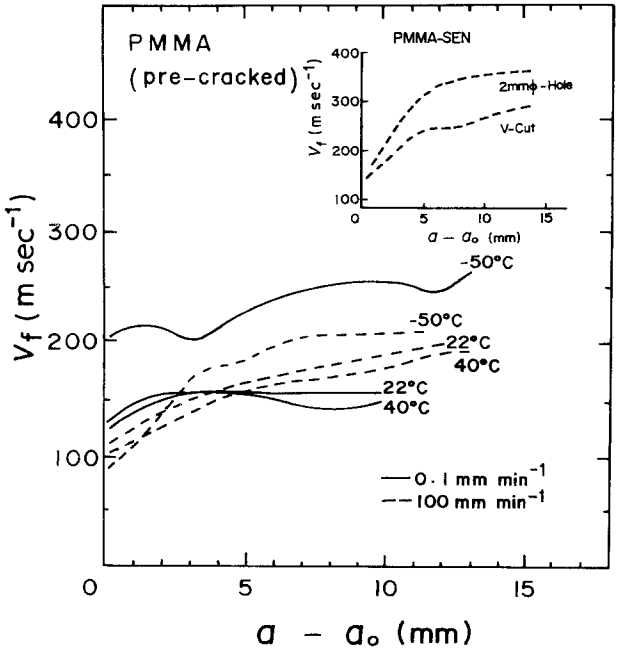

Figure 4 Results of the v_f measurement for variously notched PMMA tensile specimens.

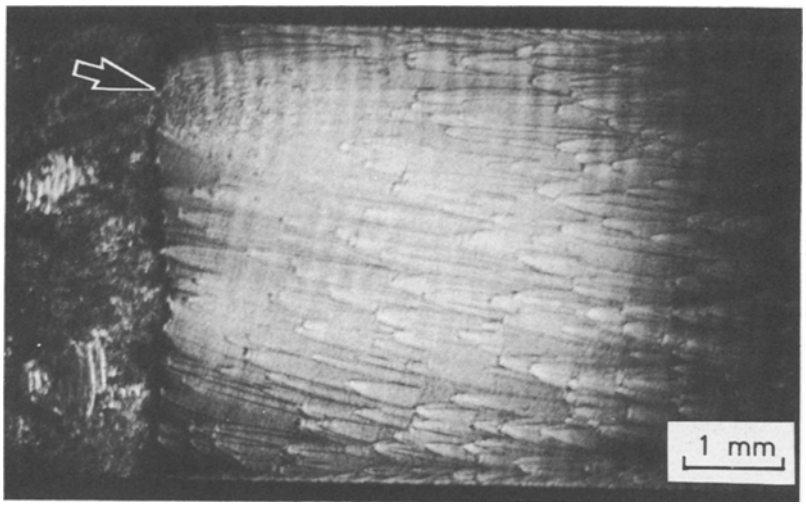

Figure 5 Fracture surface of a PMMA tensile specimen loaded at a cross-head speed of 0.1 mm min^{-1} under a temperature of $-50°$ C. An arrow indicates the exact position where the instability took place.

of 40, 22 and $-50°$ C. For every testing condition employed in Fig. 4 two specimen pieces were prepared very carefully and tested. Because the measured velocity change in each of the specimens was similar, in order to avoid confusion, the result of an arbitrary one was employed as a representative of the two, and is shown in the figure by a smoothed line. Although the experimental condition differs considerably, it should be noted that the initial starting velocity, v_{fc}, for all specimens in Fig. 4, but one which was pulled slowly at a temperature of $-50°$ C, falls in a very small range from 90 to 130 m sec^{-1}. This result is consistent with the result of the CT specimen which had the v_{fc} value of 120 m sec^{-1}.

If we look into detail about the initial velocity v_{fc}, a tendency is discernible: the v_{fc} for the slowly loaded specimens tends to become a little bit higher than that for rapidly loaded specimens. The situation, however, reverses itself in the propagation process. While the v_f of the former stayed almost constant after the slight increase in the initial course, the slight increase of v_f of the latter continued longer. In the exceptional case stated above the crack started from a velocity of about 200 m sec^{-1}, which is almost twice the v_{fc} of other specimens. A plausible cause for this may be extracted from morphology of the fracture surface shown in Fig. 5. The roughness in the slow crack region of this specimen is much more enhanced than that on other specimen surfaces (see, for example Fig. 2). This suggests that the multiple crazing at the tip of the crack in the slow area was promoted by the low temperature during the loading procedure of longer time. The more the number of craze layers at the crack tip, the more the accumulated deformation energy, which seems to result in a higher value of the fracture velocity after the instability. On occasion this should occur even at room temperature in a specimen which has a notch introduced by a rough procedure [15].

The effect of the notch root profile on the fast fracture initiation process was examined by adopting the single edge notch root V-shaped profile, and a drill hole of 2 mm diameter. The total length of each notch was about 8 mm. Fig. 6 shows the fractured surface of the specimen which had a single notch with a V-shape root. Fast fracture in this case initiated at a central part of the instability line with an initial velocity less than 146 m sec^{-1}. Results of the v_f measurement are also indicated in Fig. 4. It seems that the shape of the notch root has a less dominant effect on the initial velocity, although the effect is clearly seen on the propagation procedure as compared with the cases of pre-cracked specimens, which may rationally be explained by dissimilarity of microscopic deformation energy distribution near the notch root.

3.3. Bend and Charpy tests for PMMA specimens

The experimental results given in Sections 3.1 and 3.2 predict that similar results may be obtained in bend tests. This was confirmed for pre-cracked PMMA bend specimens at loading speeds of 0.1 and 100 mm min^{-1}. The results are shown in Fig. 7. The slowly loaded specimen exhibited crack arrest many times, whereas the rapidly loaded one had a long continuous crack propagation length. The

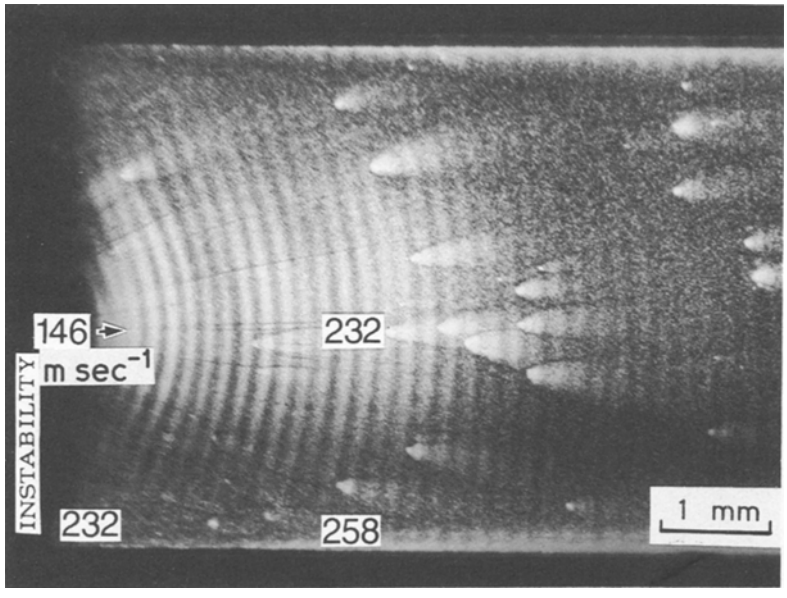

Figure 6 Fracture surface of a PMMA tensile specimen which had a single edge of the notch root of a V-shape.

value of v_{fc} for both cases was in a range from 120 to 150 m sec^{-1}, which is in accord with the results of the tensile specimens stated in the previous sections.

The effect of loading rate in bend tests was further examined by performing Charpy impact tests. Pre-cracked Charpy pieces of different height (h), 15 and 35 mm, were tested at room temperature. Fig. 8 presents an example of a fracture surface of an ultrasonically modulated Charpy specimen which had the height of 35 mm. In this case the fast crack is shown to have initiated at a point centred on the instability line. From the spacing between the first and the second ultrasonic lines a value of about 120 m sec^{-1} was obtained

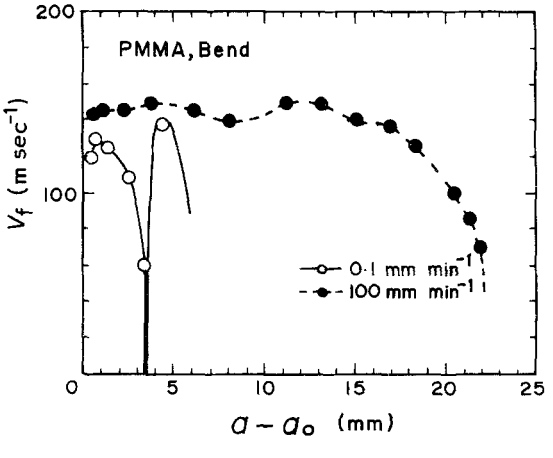

Figure 7 Results of the v_f measurement for pre-cracked PMMA bend specimens.

for the v_{fc} in this case. Results of the v_f measurement for the Charpy specimens are given in Fig. 9. The v_f of specimens of 15 mm height initially exhibited a short increase from 110 to 120 m sec^{-1}, followed by an immediate decrease which was caused by a bending effect, whereas those of 35 mm height had a much longer fast fracture area. It should be noted that in all specimens values of v_{fc} were in a range 110 to 130 m sec^{-1}, average about 120 m sec^{-1}. It is very interesting that the value of v_{fc} for pre-cracked PMMA specimens exhibited constancy over a wide range of loading speed. If we take into consideration that the hammer speed at the Charpy impact is about 2.9 m sec^{-1}, the macroscopic deformation rate at the impact may be more than 10^6 times as high as that in the static bending test above stated. The results of Figs. 2, 4, 7 and 9 imply that the value of v_{fc} of PMMA is a kind of material constant, although the constancy does not hold at lower specimen temperature or in other cases when multiple crazing/cracking at the crack tip is promoted. In the latter case, much deformation energy may be accumulated in a local area surrounding the main crack tip by the time when the instability takes place, which should yield a higher value of v_{fc}.

The physical meaning of the constancy of the v_{fc} may be closely associated with the physical meaning of the instability onset. Quite different mechanisms have been proposed to explain the mechanism of the instability onset in PMMA,

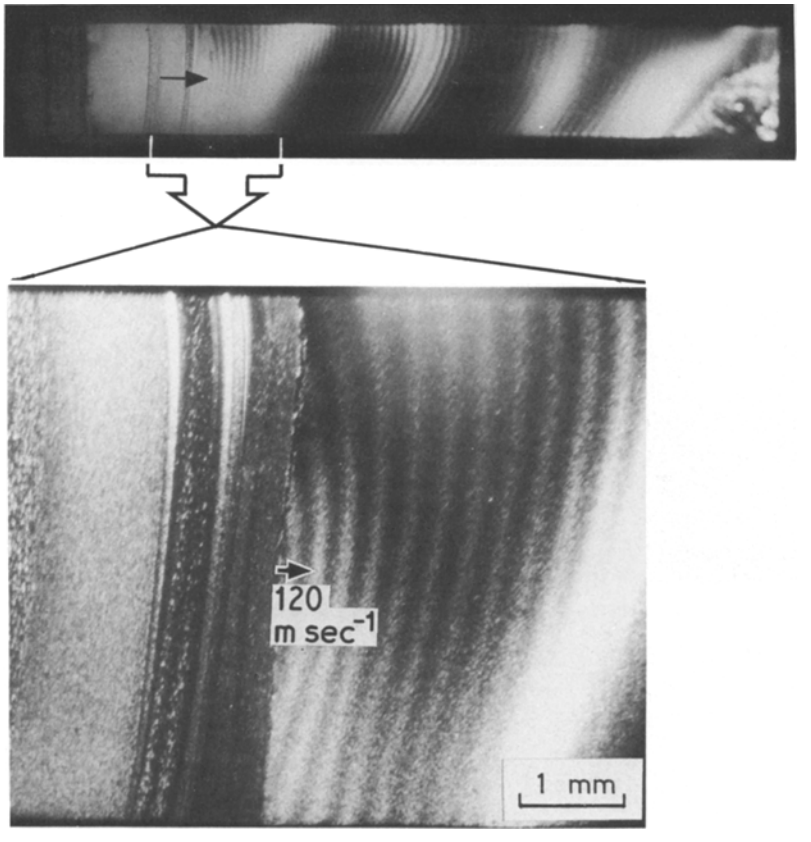

Figure 8 Fracture surface of a PMMA Charpy specimen.

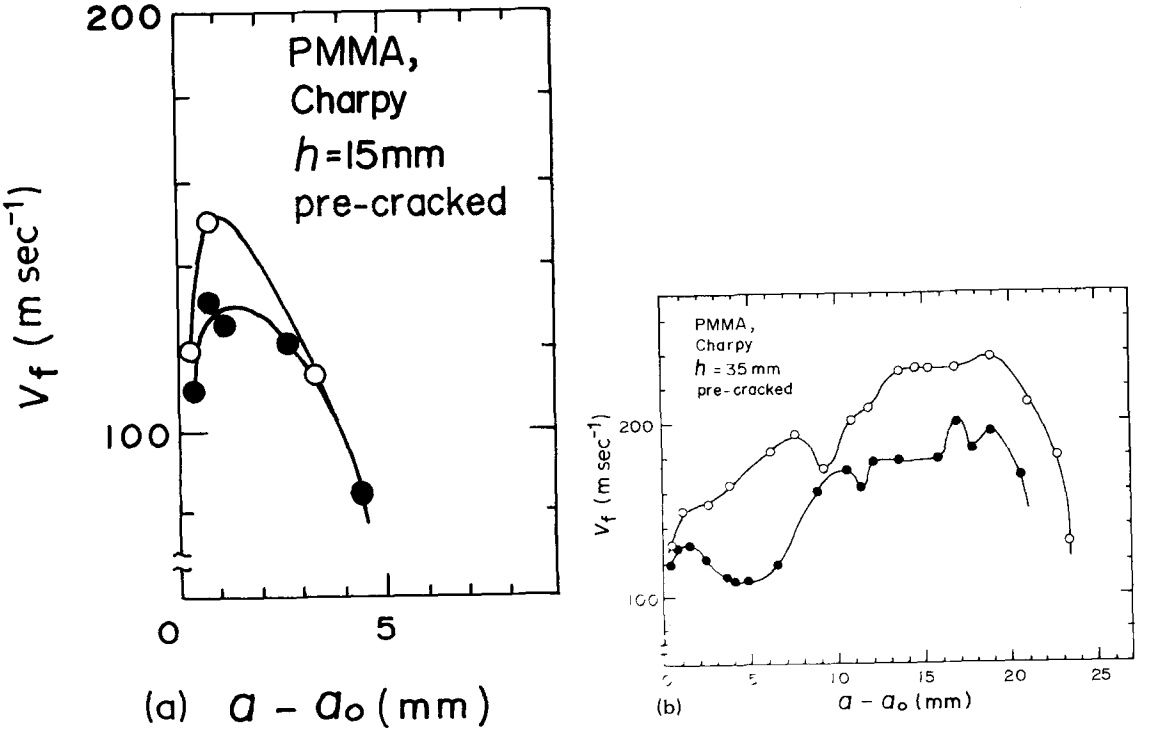

Figure 9 Results of the v_f measurement for pre-cracked PMMA Charpy specimens.

β relaxation mechanism [2] and isothermal–adiabatic transition mechanism [1]. The marked feature of the instability which has been shown by the present experiments is that the transition is highly discontinuous. Neither values below 80 m sec^{-1} for the v_{fc} of PMMA specimens in such various geometries as CT, SEN, bend and Charpy types, nor temporal fracture velocity decrease in the transition process have been obtained. This would suggest that the molecular relaxation mechanism may be hard to explain the transition phenomena, because the molecular energy absorbing mechanism should yield much more continuous velocity change. On the other hand, if we assume that the instability occurs when the slow fracture gains some critical velocity, whose existence is predicted by the isothermal–adiabatic model, the independency of the v_{fc} on the loading speed may be interpreted. Because the strain rate at the advancing crack tip is dominated rather by the crack speed than by the loading speed, the amount of local deformation energy at the crack tip just before the instability onset should not differ so much from specimen to specimen, regardless of the loading speed. Release of this similar local deformation energy should yield a similar value of v_{fc} at the instability initiation point. The initiated fast crack front should sweep away the slow crack front, forming an instability line. Naturally a faster loading speed should hasten the occurrence of the instability, making the slow crack length shorter, which is in accord with the experimental result. The temperature change from -50 to 40° C seems to have had an effective influence on the value of v_{fc}, probably because the strain rate at the crack tip had the more dominant effect, although the situation is changed when specimens are loaded slowly under low temperature, as is stated above.

Some oscillatory behaviour seen in the fracture velocity change in Fig. 9b seems to be caused by the flexural oscillation of the impacted specimen. Detailed experimental results about the flexural oscillation of PMMA Charpy specimens are given in [19].

3.4. Tensile tests for single edge notched low molecular weight PMMA specimens

It has been shown that molecular weight has considerable influence on the morphology of a PMMA fracture surface [11, 20]. This indicates that molecular structure should have an influence on the crack propagation procedure. In order to see the effect of molecular weight on the present measurement, the acrylic low molecular weight PMMA specimens, P(MMA/MA), were also adopted in the present study. The static mechanical properties such as the breaking stress, the breaking strain and Young's modulus are almost similar for both materials, whereas the Izod impact strength of P(MMA/MA) is a little, about 10%, inferior to that of the higher molecular weight PMMA material employed. Results of the v_f measurement are shown in Fig. 10. It is shown that the value of v_{fc} is not influenced by the present molecular weight change. The value is again 120 m sec^{-1} on average. The specimen loaded at the rate of 0.1 mm min^{-1} at -50° C again behaved exceptionally. On the crack propagation procedure, however, we can see effects of loading rate and of temperature as compared with the case of higher molecular weight specimens (see Fig. 4). Those effects may be caused by the molecular structure of the material; the lower molecular weight material should have less molecular chain entanglement resistance against fracturing stress. However, this resistance seems to depend on the strain rate. If we take into consideration that the stress–strain curves of this material, obtained dynamically at an elongation rate of 100 mm min^{-1} and statically at 0.1 mm min^{-1}, are similar to those of higher

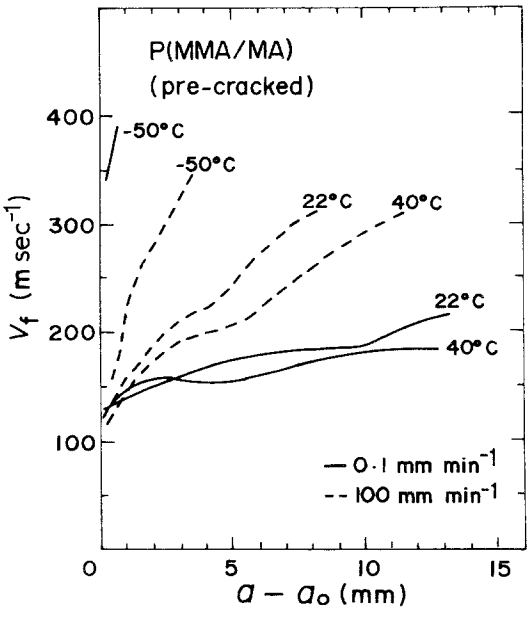

Figure 10 Results of the v_f measurement for pre-cracked P(MMA/MA) tensile specimens.

molecular weight PMMA, it may be said that the effect of molecular weight manifests itself at such a very high strain rate as in the impactive fracture process.

4. Conclusions

Ultrasonic fractography is applied to acrylic polymers with different molecular weight. Various specimen geometries were adopted including single edge notched, compact tension, bend and Charpy types. Fast fracture at the slow-to-fast transition initiates from a point source, whereas it is arrested almost one-dimensionally. The transition is shown to be highly discontinuous. No signs of gradual velocity increase are observed near the instability point. The initial velocity evaluated by the present method has been shown to fall in a small range, 90 to 150 m sec^{-1}, independent of the loading speeds (0.1 and 100 m sec^{-1}), of specimen temperature (from -50 to 40° C) and of the molecular weight change of one figure, with exceptional cases where tensile specimens were loaded slowly at a low temperature. Effect of the molecular weight has been shown only in the fast propagation procedure. The fracture velocity is more easily accelerated in this material.

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