THE FRACTURE TOUGHNESS OF POLYTETRAFLUOROETHYLENE

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Abstract

Polytetrafluoroethylene (PTFE) is an important structural polymer that in the "7C" derivative is used for gaskets, bearing pads, piston rings and diaphragms. Addition of 10 to 40 weight % aluminum 5:m spheres can increase stiffness and strength while the effect on toughness can be either positive or negative, depending on the loading rate and test temperature, as this polymer is very viscoelastic.

In this study compact tension (C(T)) specimens of pure PTFE and two aluminum filled mixtures have been tested at temperatures from 33^{o}C to –10^{o}C at crack mouth opening displacement rates from 2.5 x10^{-6} mm/s to 350 mm/s. Previous methods used to obtain fracture toughness for polymers have involved multi-specimen methods [1-5], but these were not feasible for this study because the material availability was limited and many different test conditions were to be investigated. The normalization method originally proposed by Landes and co-workers [6-7] and subsequently included in Annex A15 of ASTM E1820-99a[8] (Standard Test Method for Measurement of Fracture Toughness), has been adapted[9] for use with this polymeric material to obtain J_{Ic} and J resistance curves directly from the measured load versus crack mouth opening displacement records. Work of Bernal and associates[10-12] has shown that a load separation criterion is valid for polymeric materials using the methods of Sharobeam and Landes [13]. This work and work by Che et al.[14] has also demonstrated the equivalence of normalization function methods and multi-specimen methods at test conditions where both methods can be used.

J resistance curve results show the fracture toughness varies widely over the narrow range of test temperatures investigated here, while the mixture effects, orientation effects, and rate effects are much less dramatic. A brittle run/arrest phenomena similar to brittle "pop-ins" often observed in metals or weldments is observed at higher loading rates in all mixtures, while a thermal creep type crack growth appears important for the mixture including 25% aluminium.

Background

The U.S. Navy has become increasingly reliant on polymers in weapons systems and this has resulted in a desire to characterize interactions between polymer properties associated with strain rate sensitivity, pressure sensitivity, yield surface, failure mechanisms, damage mechanisms, spall strength, crystalline response to shock loading, and dynamic fracture mechanics. Polytetrafluoroethylene (PTFE) is an important structural polymer that in the "7C" derivative is used for gaskets, bearing pads, piston rings and diaphragms. Addition of 10 to 40 weight % aluminum 5:m spheres can increase stiffness and strength while the effect on toughness can be either positive or negative depending on the loading rate and test temperature as this polymer is very viscoelastic. Tensile and fracture properties of the PTFE

material have been reported in references [9, 15-16], while similar measurements obtained on the PTFE/aluminium mixture are available in [17-18].

Experimental Details

PolyTetraFluoroEthylene is a semi-crystalline polymer. The filled PTFE material of interest here is obtained by adding either 10% or 25% by weight of pure aluminum powder containing 5 ± 2 :m spheres to PTFE 7C powder and then preforming under pressure into a cylindrical puck with a diameter of 89 mm and a thickness of approximately 39 mm. Finally the puck was sintered using a precise heating and cooling cycle to consolidate the material. The resulting aluminum filled PTFE material has properties which are dominated by the PTFE 7C matrix material, but which is stronger, less flexible, and less fracture resistant than the PTFE material alone. The pure PTFE derivative tested here was prepared using the same performing and sintering procedure, but without adding the aluminium powder spheres.

The specimen geometry used in this study was a ½T compact tension (C(T)) geometry. The specimens were cut from the PTFE pucks in two orientations, as shown in Figure 2 and denoted as (1-2) and (2-1), with the first digit being the direction perpendicular to the crack plane and the second digit being the direction of intended crack extension. The notch tip was machined using a sharpened slitting saw and subsequently sharpened using a razor blade forced into the notch to a prescribed notch depth.

Fracture toughness was measured in this work, using standard ASTM E1820-99a compact fracture toughness specimens that were machined from the sintered pucks. Specimens were tested with two crack orientations, using three test temperatures, and four to six test rates to determine if fracture would occur in this material, and if so, how the fracture toughness depends on the orientation, test temperature, and specimen loading rate. Because the material available to test was limited, the multiple specimen test method, the "basic" method in ASTM E1820, was not considered to be an acceptable test procedure, and a single specimen test procedure was sought for obtaining fracture toughness properties for the PTFE 7C and aluminum filled PTFE polymer material. Also since it was desired to develop fracture

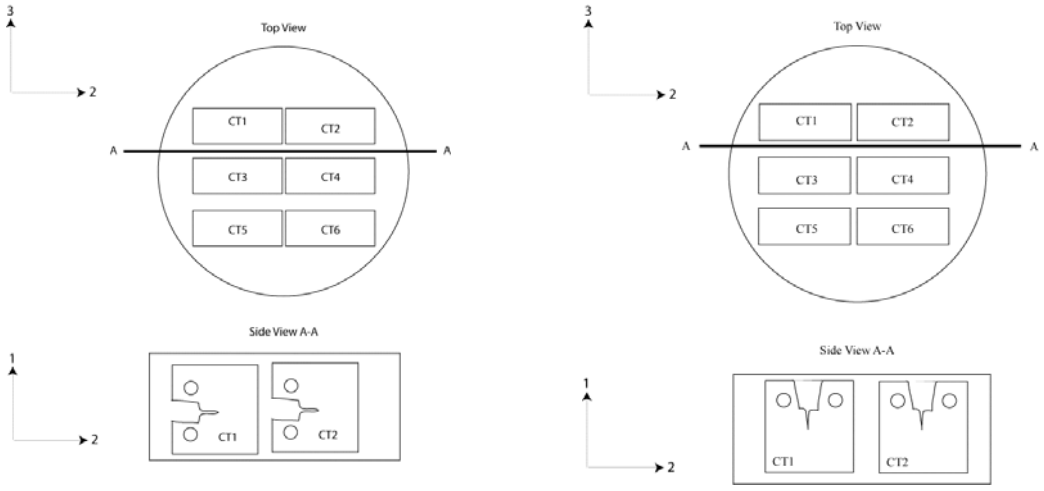


Figure 1 Specimen and cutting diagrams showing (1-2) and (2-1) orientations.

toughness properties over a range of test rates and temperatures it was felt that the number of specimens that would have to be tested to use the multi-specimen method was prohibitive.

All tests were conducted in a 100kN servohydraulic test machine with a 5kN load cell using loading rates from 360 mm/s to 2.5 x 10^{-4} mm/s. The specimen was basically ramped to a final displacement of between 2 and 5 mm, the specimen was then unloaded using the manual "set point" control, and then removed from the test fixtures. The 5 mm final displacement was used on specimens that were expected to behave in a tough fashion, while the lesser final displacements were used for higher rate or colder test conditions where extensive crack extension was expected, crack extension that would be beyond the analysis capability of the normalization method. Rapid tests took a few milliseconds, while the slowest tests took approximately 6 hours. Tests were conducted in an environmental chamber at test temperatures of -10°C, 10°C, 22°C, and 33°C. Load, crack mouth opening displacement (CMOD), actuator displacement and elapsed time were measured. CMOD was measured using a clip gage installed on integral knife edges on the load line as is typical for J integral measurements according to ASTM E1820.

Once fracture tests were completed the extent of crack growth was marked using a blue "lay-up" dye, then the specimens were reloaded to failure by dynamic crack growth after immersion in liquid nitrogen. Crack lengths and crack extensions were measured from fracture surface photographs using a digitizing tablet system.

Analysis

The basic input data consists of a load, displacement, time record for the test, a measured initial crack length, a_i, and a measured final crack length, a_f. The load is normalized according to:

$$P_{Ni} = \frac{P_i}{WB\left[\frac{W - a_{bi}}{W}\right]^{\eta_{pl}}} \tag{1}$$

where a_{bi} is the blunting corrected crack length at the ith data point and $\eta_{pl} = 2 + 0.522(W-a_{bi})/W$, with W the width of the fracture specimen. The plastic (inelastic) component of the load line displacement is normalized as:

$$CMOD'_{pl} = CMOD_{pl} / W \tag{2}$$

A normalized load displacement record for a PTFE ½T C(T) specimen is shown in Figure 2. The initial tangent stiffness measured from the load displacement record and the measured specimen crack length were used in the compliance relationship of ASTM E1820 to define an effective elastic modulus for the specific loading rate to allow the separation of the elastic and "plastic" or inelastic components of the total *CMOD* to obtain $CMOD_{pl}$. Because this material is very rate sensitive, the initial elastic modulus obtained from tensile tests could not be used to separate the elastic and plastic components of CMOD for the fracture tests. The effective modulii used for each fracture test was obtained from the individual test record and is included in Table 1. Figure 2 shows the full data set normalized based on the initial crack length with the exception of the final data point, shown as a triangle, which was normalized using the optically measured nine-point average final crack length. A reduced data set is then

obtained including data short of maximum load, and if possible, short of the onset of ductile crack initiation. This data is adjusted by using an blunting corrected crack length.

A normalization function of the form:

$$P_N = \frac{a + b*CMOD'_{pl} + c*CMOD'^{\,2}_{pl}}{d + CMOD'_{pl}} \tag{3}$$

is then fit to the reduced set of normalized data for a typical specimen, specimen PTFE58, as shown in Fig. 2. The full data set is now analyzed to determine the crack length required to place each load displacement pair on the normalization function of Eq. 3. Associating each *CMOD*-load pair with a correct crack length between the initial and final optically measured crack lengths will place the data point on the normalization function. The crack lengths required provide the crack extension that is taking place during the course of the test.

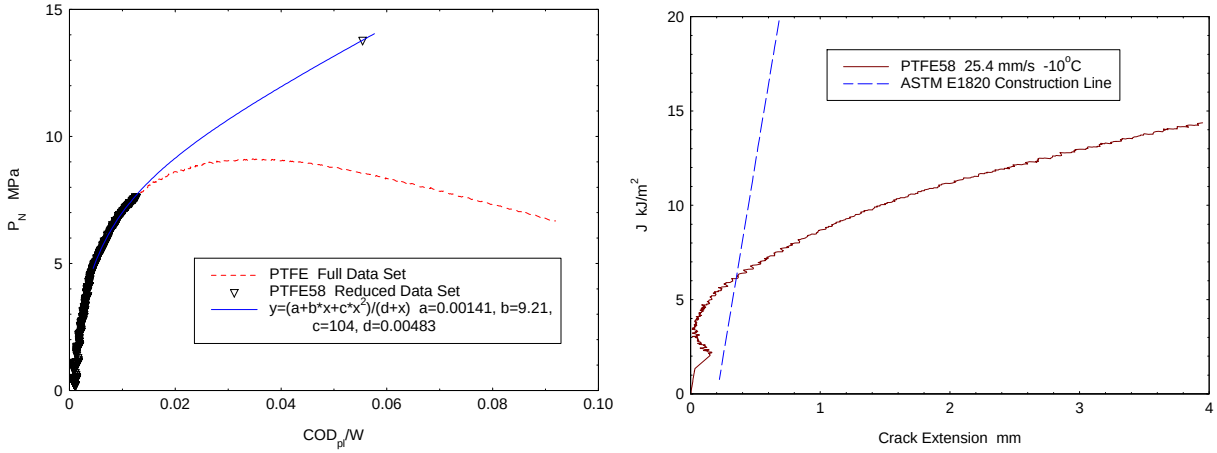


Figure 2 Example normalization function fit and resulting J-R curve.

The result is that each load displacement pair can be associated with a crack extension, and the data necessary to obtain a *J-R* curve according to ASTM E1820 is determined, with the J-R curve for being recorded during each test. The resulting J-R curve for specimen PTFE58 is shown in Fig. 2. After completion of the analysis, load, CMOD, J, crack extension and elapsed time data is available, with generally between 1000 and 2000 data sets.

Discussion of the Results

This program has investigated three compositions of the PTFE/aluminium polymer, four test temperatures, seven loading rates and two orientations, and the complete set of results can not be presented here. Most of the work has been presented for in Joyce[9] and Joyce and Joyce[16-17] including tabulated results and J-R curves for the 100+ tests completed to date. In this document an attempt is made to present a broad overview of the results. Figure 3 shows J-R curves for several specimens taken from the 25% composition data set since this data set shows more clearly all the different material behaviours that have been observed in these polymers. Each result is identified in this figure with a specimen number and a test loading rate in mm/s. Specimens tested at normal loading rates, i.e. between 0.0254 mm/s and 2.54 mm/s, tended to follow the ASTM E1820 “blunting/construction” line, resulting in at most 0.5 mm of crack extension while undergoing a rather large 5 mm of crack mouth

opening displacement (CMOD). Specimens tested at 25.4 mm/s demonstrated small unstable run/arrest events which here have been called “pop-ins”. The specimens that were tested at approximately 360 mm/s resulted in smooth, apparently “ductile” J-R curves. All of these tests demonstrated very little effect of the specimen orientation. The very slow tests conducted here, however, at the 0.00025 mm/s loading rate, which corresponds to about a 6 hour ramp to a CMOD = 5 mm, showed extensive crack extension, with the (1-2) orientation resulting in much more extensive crack extension at lower J integral levels than that found in the (2-1) orientation. The tests at 0.0025 mm/s resulted in only crack tip blunting for the (2-1) orientation (specimen 6), while the corresponding (1-2) test resulted in more than 3 mm of crack extension (specimen 52).

The viscous crack tip blunting behaviour at normal testing rates was as expected, but the mechanism behind the extensive “creeping” crack growth that was observed at the very slow rates is not understood.

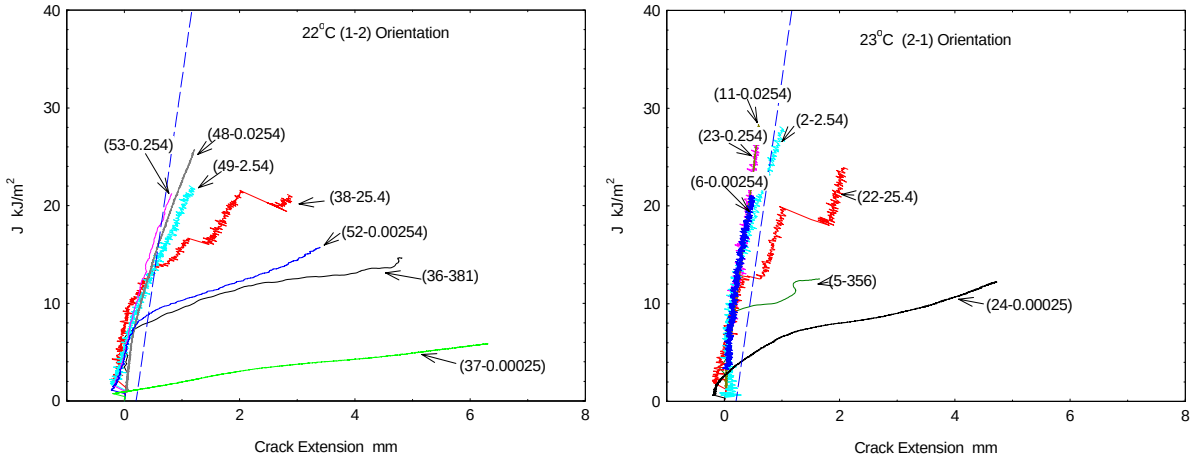


Figure 3 25% composition showing a) (1-2) and b) (2-1) orientation results at 23°C.

Figure 4 combines results obtained from the 0% (PTFE), 10%, and 25% compositions that were run at lower loading rates and higher temperatures. Figure 4 shows that the creep crack growth was much more predominant in the 25% aluminium composition than in either the 10% aluminium or pure PTFE compositions and is quite possibly associated with the aluminium/PTFE interfaces, which are expected to be very low in strength. Specimens are designated as PTFE, “-10” or “-25” according to their aluminium compositions.

Figure 5 shows a comparison of fracture surfaces obtained from a region of a) crack tip blunting and b) creep crack growth. The 5μm aluminium spheres are clearly more exposed during the creep crack than during the ductile crack tip blunting.

Run/Arrest Pop-ins

Typical load versus CMOD test records and the corresponding J-R curves are shown for three PTFE samples in Figure 6. Corresponding fracture surface photographs are shown in Figure 7. There is a clear one-to-one correspondence between the run/arrest “pop-in” events and the markings on the fracture surfaces. Pop-ins are not observed on very slowly loaded specimens or specimens tested at elevated temperature for which the viscoelastic relation rate was sufficient to remove this effect. High rates of loading appeared to effectively outrun the run/arrest events. Pop-ins are observed at intermediate loading rates and at intermediate

temperatures for all compositions and both orientations. The most complete set of results is presented for the PTFE composition in Figure 8 which shows the transition from blunting at slow loading rates to large pop-ins, to continually smaller pop-ins, to smooth J-R curves as

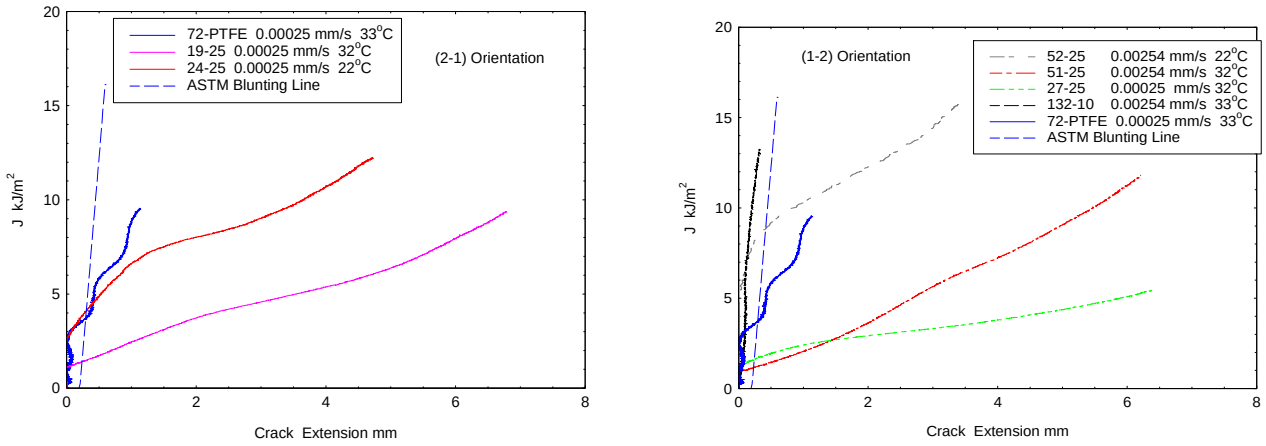
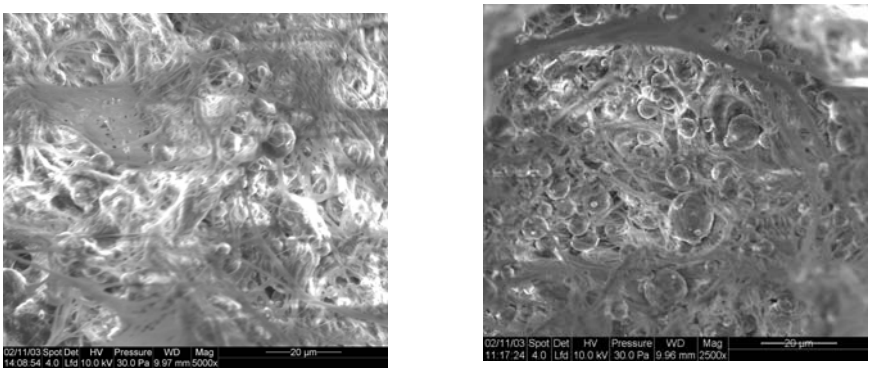


Figure 4 Slow loading rate creep crack growth predominates in the 25% composition.



(a) (b)

Figure 5 SEM photographs of (a) crack tip blunting and (b) creep crack growth regions for 25% composition specimens.

the test loading rate is increased. While it seems quite intuitive that run/arrest behaviour would develop as the loading rate is increased beyond what the viscoelastic relaxation can accommodate, it is surprising that stable, ductile-like behaviour is again established at loading rates exceeding 100 mm/s. A summary of high loading rate J-R curves for the three compositions at the three higher temperatures is presented in Figure 9. Attempts to test at higher loading rates have been unsuccessful to date with the load displacement records becoming too oscillatory to analyze successfully with the normalization function technique.

Conclusions

Testing this polymer using multi-specimen procedures at standard laboratory testing rates and ambient temperatures would result in missing most interesting features. Use of the normalization procedure allows observation of the complex transition from creep-crack-growth behaviour, to viscous blunting, through the run/arrest or pop-in behaviours, to the smooth ductile-like J-R curve behaviour observed here only at the higher loading rates and/or

higher test temperatures. Developing a material model that incorporates all of these effects is still a work in progress, but a technique to measure the polymer behaviour is now available.

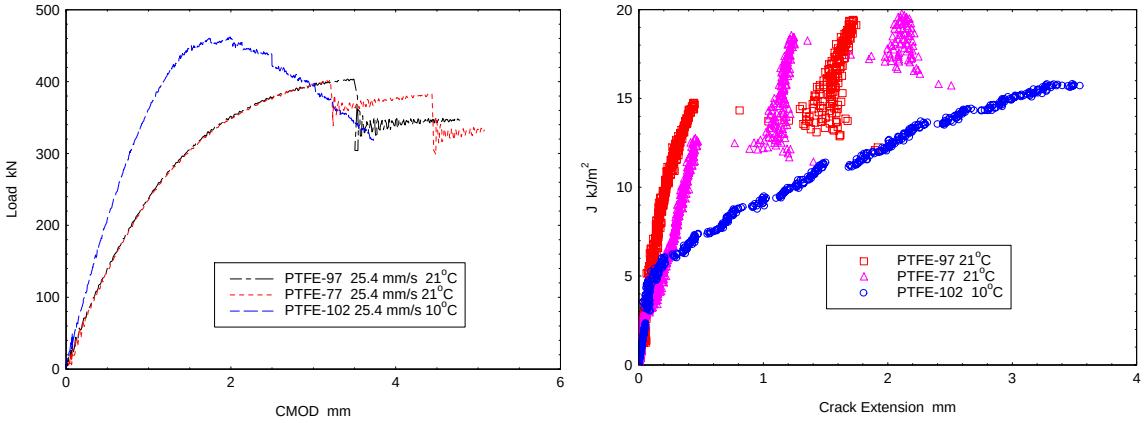


Figure 6 Load versus CMOD and J resistance curves demonstrating "pop-in" behaviour.

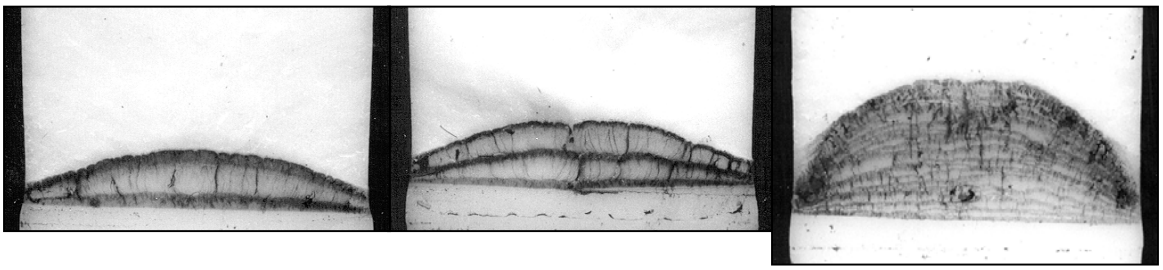


Figure 7 Run-arrest fracture surfaces of PTFE specimens 97, 77, and 102 respectively.

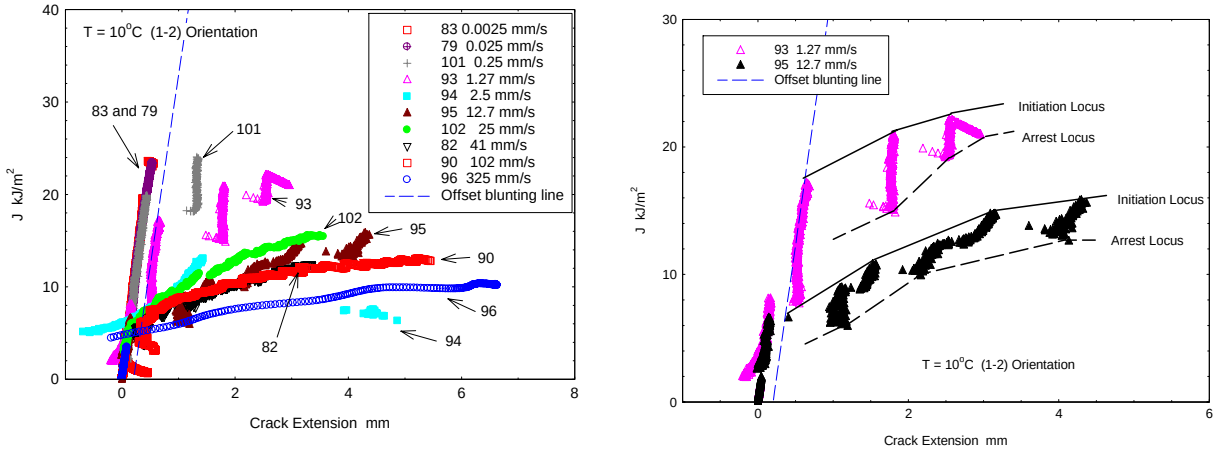


Figure 8 Pop-in behaviour for the PTFE polymer over a range of loading rates.

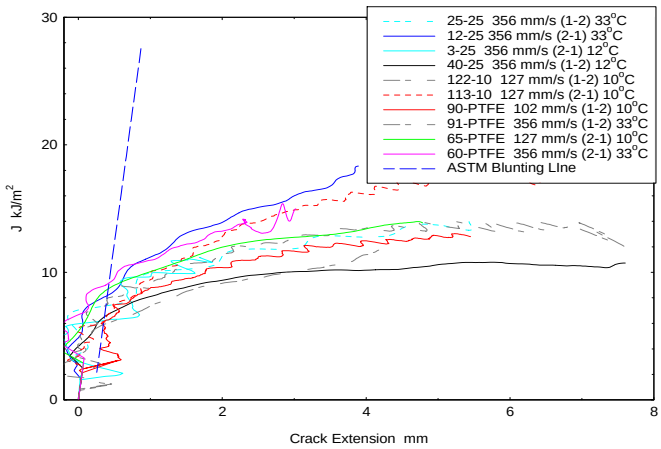


Figure 9 J-R curves for high rate tests showing ductile-like crack extension and little dependence on composition, loading rate, or orientation.

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